

# PATENT ABSTRACTS OF JAPAN

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(71)Applicant : DAINIPPON INK & CHEM INC

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(72)Inventor : IMAMURA AKIYUKI

TAKAHASHI AKIO

ARIGA TOSHIRO

MATSUKI KEIKO

KAMIKURA MASAO

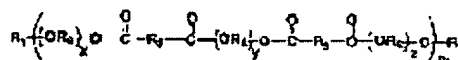
## (54) PRODUCTION OF LACTIC-ACID-BASED POLYETHER POLYESTER

### (57)Abstract:

**PURPOSE:** To provide a biodegradable lactic-acid-based copolymer having a sufficiently high molecular weight, sufficient heat resistance and sufficient heat stability and having rigidity, flexibility and transparency suitable for the object of use by copolymerizing a specified polyether ester with a lactide in the presence of a polymerization catalyst.

**CONSTITUTION:** A polyether polyester comprising repeating units represented by formula I [wherein m, x, y, and z are each an integer of 1 or greater; R1 and R2 are each H or a hydrocarbon group; R3 and R5 are aliphatic hydrocarbon groups provided that the total of their methylenic carbon atoms is 2 or above; R2, R4 and R6 are each alkylene or a chemical structure of formula II

(wherein R8, R9 and R10 are each alkylene; a, b and c are each 0 or an integer of 1 or greater; and a+b+c is an integer of 1 or greater)] is copolymerized with a lactide (B) in the presence of a polymerization catalyst (C) to obtain a high-molecular-weight biodegradable lactic-acid-based copolymer having strengths high enough to be used as a general-purpose resin, and molding heat stability and having rigidity, transparency and flexibility suitable for the



II

purpose of use.

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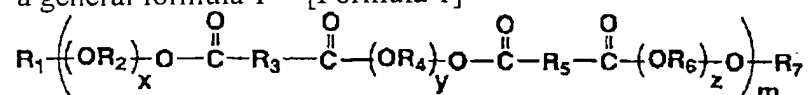
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## CLAIMS

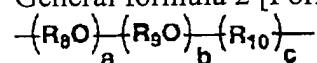
[Claim(s)]

[Claim 1] the polyether polyester (A) which has the repeat unit expressed with a general formula 1, and a general formula 1 -- [Formula 1]



(mx, y, and z are one or more integers among a formula.) R1 and R7 -- hydrogen or a hydrocarbon group, and R3 and R5 -- the sum total of a methylene carbon number -- two or more aliphatic hydrocarbon radicals -- it is -- R2, R4, and R6 -- an alkylene group -- and -- or it has the chemical structure of a general formula 2.

General formula 2 [Formula 2]



(Among a formula, R8, R9, and R10 are alkylene groups, and c is a, b, 0, or one or more integers, and a+b+c is one or more integers.)

The manufacture approach of the lactic-acid system polyether polyester characterized by carrying out copolymerization of the lactide (B) under existence of a polymerization catalyst (C).

[Claim 2] The manufacture approach of the lactic-acid system polyether polyester according to claim 1 characterized by polyether polyester (A) being polyether polyester by which macromolecule quantification was carried out by the polyvalent carboxylic acid (and/or, the acid anhydride) (D) of three or more organic functions.

[Claim 3] The manufacture approach of lactic-acid system polyether polyester according to claim 1 or 2 that polyether polyester (A) is characterized by the mole ratios of the ether group in polyether polyester and an ester group being 999 / 1 - 200/800.

[Claim 4] The manufacture approach of the lactic-acid system polyether polyester any one publication of three from claim 1 to which the weight ratio of polyether polyester (A) and lactide (B) is characterized by being 60 / 40 - 5/95.

[Claim 5] polyether polyol (E), polyol (F), and dicarboxylic acid (G) -- and/or, the manufacture approach of the polyether polyester (H) obtained by making the polyvalent carboxylic acid of three or more organic functions, and/or its acid anhydride (D) react, and the lactic-acid system polyether polyester characterized by carrying out copolymerization of the lactide (B) under existence of a polymerization catalyst (C).

[Claim 6] The manufacture approach of lactic-acid system polyether polyester according to claim 5 that polyether polyester (H) is characterized by the mole ratios of the ether group in polyether polyester (H) and an ester group being 999 / 1 - 200/800.

[Claim 7] The manufacture approach of lactic-acid system polyether polyester according to claim 5 or 6 that the weight ratio of polyether polyester (H) and lactide (B) is characterized by being 60 / 40 - 5/95.

[Claim 8] The manufacture approach of the lactic-acid system polyether polyester any one publication of seven from claim 5 characterized by polyether polyol (E) being one or more kinds of polyether polyols chosen from a polyethylene glycol, a polypropylene glycol, and the Pori (oxyethylene-oxypropylene) glycol that is the copolymer of ethyleneoxide and propylene oxide.

[Claim 9] The manufacture approach of the lactic-acid system polyether polyester any one publication of seven from claim 5 characterized by polyol (F) being one or more kinds of diols chosen from ethylene glycol and propylene glycol.

[Claim 10] The manufacture approach of the lactic-acid system polyether polyester any one publication of seven from claim 5 characterized by dicarboxylic acid (G) being one or more kinds of dicarboxylic acid chosen from a succinic acid, an adipic acid, a sebacic acid, its acid anhydride and those methyl ester, and ethyl ester.

[Claim 11] polyol (F) and dicarboxylic acid (G) -- and/or, the manufacture approach of the polyester which consists of the polyvalent carboxylic acid of three or more organic functions, and/or its acid anhydride (D), the polyether polyester (I) obtained by carrying out the ester exchange reaction of the polyether polyol (E), and the lactic-acid system polyether polyester characterized by carrying out copolymerization of the lactide (B) under existence of a polymerization catalyst (C).

[Claim 12] The manufacture approach of lactic-acid system polyether polyester according to claim 11 that polyether polyester (I) is characterized by the mole ratios of the ether group in polyether polyester and an ester group being 999 / 1 - 200/800.

[Claim 13] The manufacture approach of lactic-acid system polyether polyester according to claim 11 or 12 that the weight ratio of polyether polyester (I) and lactide (B) is characterized by being 60 / 40 - 5/95.

[Claim 14] The manufacture approach of the lactic-acid system polyether polyester any one publication of 13 from claim 11 characterized by polyether polyol (E) being one or more kinds of polyether polyols chosen from a polyethylene glycol, a polypropylene glycol, and the Pori (oxyethylene-oxypropylene) glycol that is the copolymer of ethyleneoxide and propylene oxide.

[Claim 15] The manufacture approach of the lactic-acid system polyether polyester any one publication of 13 from claim 11 characterized by polyol (F) being one or more kinds of diols chosen from ethylene glycol and propylene glycol.

[Claim 16] The manufacture approach of the lactic-acid system polyether polyester any one publication of 13 from claim 11 characterized by dicarboxylic acid (G) being one or more kinds of dicarboxylic acid chosen from a succinic acid, an adipic acid, a sebacic acid, its acid anhydride and those methyl ester, and ethyl ester.

[Claim 17] The manufacture approach of the polyether polyester (J) obtained by making the polyvalent carboxylic acid (and/or, the acid anhydride) of three or more organic functions react to polyol (F), the polyester which dicarboxylic acid (G) is made to react and is obtained, and the polyether polyester which polyether polyol (E) is made to react and is obtained, and the lactic-acid system polyether polyester characterized by carrying out copolymerization of the lactide (B) under existence of a polymerization catalyst (C).

[Claim 18] The manufacture approach of lactic-acid system polyether polyester according to claim 17 that polyether polyester (J) is characterized by the mole ratios of the ether group in polyether polyester and an ester group being 999 / 1 - 200/800.

[Claim 19] The manufacture approach of lactic-acid system polyether polyester according to claim 17 or 18 that the weight ratio of polyether polyester (J) and lactide (B) is characterized by being 60 / 40 - 5/95.

[Claim 20] The manufacture approach of the lactic-acid system polyether polyester any one publication of 19 from claim 17 characterized by polyether polyol (E) being one or more kinds of polyether polyols chosen from a polyethylene glycol, a polypropylene glycol, and the Pori (oxyethylene-oxypropylene) glycol that is the copolymer of ethyleneoxide and propylene oxide.

[Claim 21] The manufacture approach of the lactic-acid system polyether polyester any one publication of 19 from claim 17 characterized by polyol (F) being one or more kinds of diols chosen from ethylene

glycol and propylene glycol.

[Claim 22] The manufacture approach of the lactic-acid system polyether polyester any one publication of 19 from claim 17 characterized by dicarboxylic acid (G) being one or more kinds of dicarboxylic acid chosen from a succinic acid, an adipic acid, a sebacic acid, its acid anhydride and those methyl ester, and ethyl ester.

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**DETAILED DESCRIPTION**

[Detailed Description of the Invention]

[0001]

[Industrial Application] This invention relates to the manufacture approach of the lactic-acid system copolymer of the amount of macromolecules in which various fabrication useful into shaping resin, a sheet film material, coating resin, ink resin, adhesives resin, the resin for laminations to paper, and a foaming resin ingredient is possible.

[0002] It is lactide system polyether polyester of the amount of giant molecules, and the lactic-acid system copolymer made by the manufacture approach of this invention has biodegradability, and the fabrication by various approaches, such as blow molding, extrusion molding, injection molding, inflation molding, laminate molding, press forming, and extrusion-foaming shaping, is possible for it, and it can fabricate it using the shaping equipment for general-purpose resin.

[0003] The lactic-acid system copolymer made by the manufacture approach of this invention specifically As above-mentioned various the application for sheets, for example, object, to a tray, a cup, a pan, a blister, etc., as a film As a blow molding article, it is [ material / a disposable diaper, sanitary items, an artificial kidney, a suture, / medical-application ] [0004] as sanitary goods to a shampoo bottle, a cosmetics bottle, a drink bottle, an oil container, etc. in bags, such as a wrap film, food packing, other general packages, a garbage bag, a register bag, a general specification bag, and a heavy bag. As agricultural materials, into the multifilm for agriculture, the gradual release agent sheet of agricultural chemicals, an anti-bird network, a care-of-health sheet, a plant pot, a fruits bag, etc., as fishery materials To a fishing net, a dried-seaweed culture network, a fishing line, a ship bottom paint, etc., as an injection-molded product It is [0005] to the miscellaneous-goods article for days, stationery, etc., such as a shank of the heart of golf tea, false food, and a cotton swab, the rod of a candy, a brush, a gear-tooth brush, a glass syringe, a pan, a cup, a comb, and a razor, a cassette of a tape, a disposable spoon fork, and a ball-point. In addition to this as a lamination to paper, it is used for a binding tape, a prepaid card, a balloon, a panty hose, a hair cap, sponge, a cellophane tape, an umbrella, a raincoat, a plastic glove, a hair cap, a rope, a nonwoven fabric, a tube, a foaming tray, foaming shock absorbing material, shock absorbing material, packaging, hot melt adhesive, the filter of a cigarette, a T-shirt, etc. useful at a tray, a cup, a pan, a megaphone, etc.

[0006]

[Description of the Prior Art] In recent years, research which is going to utilize widely the lactic-acid system polymer which has the outstanding biodegradability from an environmental problem etc. as a general-purpose polymer is done briskly, and many researches on the manufacture approach and patent application are made. However, conventionally, if it is hard to say that the copolymer of the polylactic acid which is the polymer of a well-known lactic acid or lactide or a lactic acid, and other monomers has the sufficiently satisfactory engine performance in a moldability, transparency, and thermal resistance and a special application is removed, there is a trouble in using as general-purpose resin, and much of amelioration of these lactic-acids system polymer is expected.

[0007] The block copolymer which consists of a polylactic acid segment and a polypropylene-glycol

segment, its manufacture approach, a copolymer film, and copolymerization are indicated by JP,1-108226,A. Moreover, the copolymer of a lactic acid and a polyethylene glycol is indicated by JP,1-108226,A.

[0008] However, by these approaches, if the amount of copolymerization of polyol is made [ many ], molecular weight will become extremely low. For example, in the example 4 of JP,1-108226,A, when 10 % of the weight of polypropylene glycols of number average molecular weight 4,000 is copolymerized with lactide, as for the number average molecular weight of the copolymer obtained, only the copolymer of 30,000 is obtained. Moreover, it is difficult to lose the reinforcement of the copolymer which consists of these lactic acids and polyol, if about 40 degrees C is exceeded, and to be equal to the use in an elevated temperature as common mold goods other than medical application.

[0009] As a copolymer of lactide and aliphatic series polyester, the polymerization of the epsilon-caprolactone is carried out beforehand, a homopolymer is obtained, and the approach of block-copolymerization-izing lactide further is indicated by JP,63-145661,A.

[0010] However, by the approach of block-copolymerization-izing lactide to a Pori epsilon-caprolactone, the obtained copolymer carries out nebula opacification. It is hard to dissolve the Pori epsilon-caprolactone block and polylactic acid block in a copolymer, and this is considered for becoming cloudy reflecting the high crystallinity which the aliphatic series system polyester of a Pori epsilon-caprolactone chain generally has. Moreover, in spite of the comparatively high glass transition point by differential thermal analysis, it is a flexible property at a room temperature.

[0011] When these Prior arts were summarized and sufficient reinforcement, thermal resistance, and thermal stability were given, it lacked in flexibility and transparency, and since it would become scarce at reinforcement, thermal resistance, and thermal stability if sufficient flexibility and transparency are given, the polymer equipped with the property it can be satisfied with still using it as ingredient resin, such as a film sheet, of a property enough was not obtained.

[0012] Plasticization of the polymer by the additive can be considered as these physical-properties improvement means. However, an improvement of the physical properties by these plasticization approach also has problems, such as reduction, disappearance, etc. of the plasticization effectiveness according [ the lactide which is a plasticizer ] to disappearance, out of a polymer at exsorption and a list during storage of the adhesion contamination problem to the equipment in the production process by a heat-resistant fall and sublimation scattering of lactide, and a product, or use, when the lactide which is a residual monomer is used as a plasticizer as a means of plasticization.

[0013] Moreover, also when the plasticizer for the polymers of general well-known common use was added, a lot of plasticizer addition for plasticization was needed, the problem of the bleed out of a plasticizer is unescapable, problems, such as disappearance of the plasticization effectiveness of a under [ storage ], were not solved, and the polymer equipped with a satisfying enough property as a polymer for wrapping was not obtained. Moreover, it was not easy to obtain the lactic-acid system polymer of the amount of macromolecules, either.

[0014]

[Problem(s) to be Solved by the Invention] Therefore, the technical problem which this invention tends to solve is to offer the manufacture approach of the lactic-acid system polyether polyester of biodegradability of having sufficient amount of macromolecules, thermal resistance, and thermal stability, and having the rigidity and flexibility according to an application, and transparency.

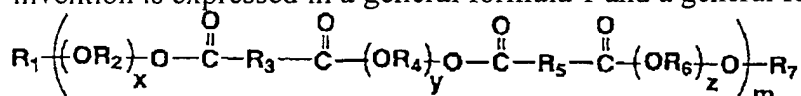
[0015]

[A means to solve a technical problem] Wholeheartedly this inventions as a result of examination that such a technical problem should be solved Lactide, By making the polyether polyester which consists of the polyether polyol component and polyol component which consist of various configuration rates, and a dicarboxylic acid component react as an indispensable component Moreover, by making the polyether polyester which consists of lactide, and the polyether polyol component and polyol component which consist of various configuration rates, a dicarboxylic acid component, the polyvalent carboxylic acid of three or more organic functions and/or its acid anhydride react as an indispensable component Controlling the decomposition to a monomer, having the molding temperature of the large range, and

having the thermal stability at the time of high reinforcement and shaping at the time of shaping, [0016] Moreover, that various lactic-acid system copolymers of even the resin which has the flexible high property of the toughness which has high molecular weight easily, tears when it is processed in the shape of a film from hard resin with a high glass transition point and the high melting point, is excellent in reinforcement, and cannot carry out a chip box crack easily changing the rate of this polyether polyester into arbitration in case copolymerization-ization is performed with lactide, or by changing the class of polyether polyester can be manufactured, [0017] That is, it finds out that the lactic-acid system copolymer of the amount of macromolecules of resolvability which has sufficient reinforcement which can be used as general-purpose resin, and the thermal stability at the time of shaping, and has the rigidity according to an application, transparency, and flexibility can be manufactured, and came to complete this invention.

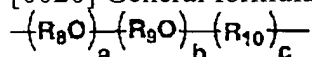
[0018]

[Elements of the Invention] namely, the polyether polyester (A) which has the repeat unit as which this invention is expressed in a general formula 1 and a general formula 1 -- [Formula 3]



[0019] (mx, y, and z are one or more integers among a formula.) As for R1 and R7, the sum totals of a methylene carbon number of hydrogen or a hydrocarbon group, and R3 and R5 are two or more aliphatic hydrocarbon radicals, and R2, R4, and R6 have the chemical structure of an alkylene group or a general formula 2.

[0020] General formula 2 [Formula 4]



(Among a formula, R8, R9, and R10 are alkylene groups, and c is a, b, 0, or one or more integers, and a+b+c is one or more integers.)

It is the manufacture approach of the lactic-acid system polyether polyester characterized by carrying out copolymerization of the lactide (B) under existence of a polymerization catalyst (C).

[0021] Moreover, the manufacture approach of the lactic-acid system polyether polyester of this invention being polyether polyester with which macromolecule quantification of this polyether polyester (A) was carried out by the polyvalent carboxylic acid (and/or, the acid anhydride) (D) of three or more organic functions, and this polyether polyester (A) are characterized by being 999 / 1 - 200/800 by the mole ratio of the ether group in polyether polyester, and an ester group.

[0022] Moreover, the manufacture approach of the lactic-acid system polyether polyester of this invention is characterized by the weight ratios of polyether polyester (A) and lactide (B) being 60 / 40 - 5/95.

[0023] furthermore, this invention -- polyether polyol (E), polyol (F), and dicarboxylic acid (G) -- and/or, it is the manufacture approach of the polyether polyester (H) obtained by making the polyvalent carboxylic acid of three or more organic functions, and/or its acid anhydride (D) react, and the lactic-acid system polyether polyester characterized by carrying out copolymerization of the lactide (B) under existence of a polymerization catalyst (C).

[0024] The manufacture approach of the lactic-acid system polyether polyester of this invention is characterized by the weight ratios of that polyether polyester (H) is [ the mole ratios of the ether group in polyether polyester (H) and an ester group ] 999 / 1 - 200/800, this polyether polyester (H), and lactide (B) being 60 / 40 - 5/95.

[0025] Furthermore, polyether polyol (E) is characterized by being one or more kinds of polyether polyols chosen from a polyethylene glycol, a polypropylene glycol, and the Pori (oxyethylene-oxypropylene) glycol that is the copolymer of ethyleneoxide and propylene oxide especially in detail. Moreover, polyol (F) is characterized by being one or more kinds of diols chosen from ethylene glycol



and propylene glycol.

[0026] Moreover, a carboxylic acid (G) is characterized by being one or more kinds of dicarboxylic acid chosen from a succinic acid, an adipic acid, a sebacic acid, its acid anhydride and those methyl ester, and ethyl ester especially in detail.

[0027] furthermore, this invention -- polyol (F) and dicarboxylic acid (G) -- and/or, the manufacture approach of the polyester which consists of the polyvalent carboxylic acid of three or more organic functions and/or its acid anhydride (D), the polyether polyester (I) obtained by carrying out the ester exchange reaction of the polyether polyol (E), and the lactic-acid system polyether polyester characterized by carrying out copolymerization of the lactide (B) under existence of a polymerization catalyst (C) is also included.

[0028] As for the manufacture approach of the lactic-acid system polyether polyester of this invention, polyether polyester (I) is further characterized by to be 999 / 1 - 200/800 and the weight ratios of polyether polyester (I) and lactide (B) being 60 / 40 - 5/95 by the mole ratio of the ether group in polyether polyester, and an ester group.

[0029] Moreover, it is that polyether polyol (E) is one or more kinds of polyether polyols chosen from a polyethylene glycol, a polypropylene glycol, and the Pori (oxyethylene-oxypropylene) glycol that is the copolymer of ethyleneoxide and propylene oxide, and [0030] in detail. Polyol's (F's)'s being one or more kinds of diols chosen from ethylene glycol and propylene glycol and dicarboxylic acid (G) are the manufacture approaches characterized by being one or more kinds of dicarboxylic acid chosen from a succinic acid, an adipic acid, a sebacic acid, its acid anhydride and those methyl ester, and ethyl ester.

[0031] Furthermore, the polyester which this invention makes dicarboxylic acid (G) react with polyol (F), and is obtained, The polyether polyester obtained by making the polyvalent carboxylic acid (and/or, the acid anhydride) of three or more organic functions react to the polyether polyester which polyether polyol (E) is made to react and is obtained (J), The manufacture approach of the lactic-acid system polyether polyester characterized by carrying out copolymerization of the lactide (B) under existence of a polymerization catalyst (C) is also included.

[0032] The manufacture approach of the lactic-acid system polyether polyester of this invention is characterized by the weight ratios of that this polyether polyester (J) is [ the mole ratios of the ether group in polyether polyester and an ester group ] 999 / 1 - 200/800, polyether polyester (J), and lactide (B) being 60 / 40 - 5/95.

[0033] Furthermore, it is that polyether polyol (E) is one or more kinds of polyether polyols chosen from a polyethylene glycol, a polypropylene glycol, and the Pori (oxyethylene-oxypropylene) glycol that is the copolymer of ethyleneoxide and propylene oxide, and [0034] in detail. Polyol's (F's)'s being one or more kinds of diols chosen from ethylene glycol and propylene glycol and dicarboxylic acid (G) are characterized by being one or more kinds of dicarboxylic acid chosen from a succinic acid, an adipic acid, a sebacic acid, its acid anhydride and those methyl ester, and ethyl ester.

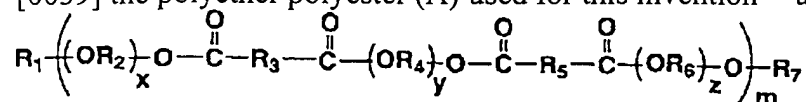
[0035] Below, this invention is explained at a detail. Sequential explanation is given about the lactide and polyether polyester which are used by this invention, dicarboxylic acid, polyether polyol, polyol, the polyvalent carboxylic acid of three or more organic functions, its acid anhydride, etc.

[0036] The lactide used by this invention is the compound which cyclic-ester-ized the lactic acid between dyad, and is a monomer which has a stereoisomer. That is, L-lactide which consists of two L-lactic acid, D-lactide which consists of a D-lactic acid, and the MESO-lactide which consists of L-lactic acid and a D-lactic acid exist in lactide.

[0037] Although the copolymer only containing L-lactide or D-lactide is crystallized and high-melting is obtained, by the manufacture approach of the lactic-acid system copolymer of the amount of macromolecules of this invention, the desirable resin property according to an application is realizable by combining these three sorts of lactide.

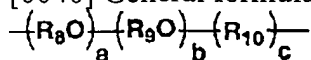
[0038] In order for the lactide used in order to discover a high thermal property in this invention to discover a thermal property with what [ desirable still higher ] contains 75% or more for L-lactide among the total lactide, as for lactide, what contains 90% or more for L-lactide among the total lactide is desirable.

[0039] the polyether polyester (A) used for this invention -- a general formula 1 -- [Formula 5]



(mx, y, and z are one or more integers among a formula.) As for R1 and R7, the sum totals of a methylene carbon number of hydrogen or a hydrocarbon group, and R3 and R5 are two or more aliphatic hydrocarbon radicals, and R2, R4, and R6 have the chemical structure of an alkylene group or a general formula 2.

[0040] General formula 2 [Formula 6]



(Among a formula, R8, R9, and R10 are alkylene groups, and c is a, b, 0, or one or more integers, and a+b+c is one or more integers.)

[0041] Furthermore, in detail, as for R1 and R7 in a general formula 1 and a general formula 2, the sum total of a methylene carbon number of hydrogen or the hydrocarbon group of carbon numbers 1-4, and R3 and R5 is the aliphatic hydrocarbon radical of 2-12, and, as for carbon numbers R2, R4, R6, R8, R9, and R10, the carbon number of an alkylene principal chain has the alkylene group of 2-15, or the chemical structure of a general formula 2.

[0042] Specifically, the hydrocarbon group more than divalent [ of methylene ethylene, trimethylene [-(CH2) 3-], propylene, tetramethylen, and [CH3CH2C(CH2-)3] [-CH(CH2-)2 grade ] especially ethylene, a propylene, and [CH3CH2C(CH2-)3] [-CH (CH2-)2 are mentioned.

[0043] the polyether polyester from which polyether polyester (H) is obtained by the dehydration and the deglycol reaction of polyether polyol, polyol, and dicarboxylic acid -- or it is polyether polyester obtained by the dehydration and the deglycol reaction of polyether polyol, polyol, dicarboxylic acid, and the polyvalent carboxylic acid (and/or, the acid anhydride) of three or more organic functions.

[0044] the polyether polyester with which the ester interchange of the polyether polyester (I) used for this invention was carried out, and polyol and dicarboxylic acid were obtained in dehydration, and the polyester and polyether polyol which is made to carry out a deglycol reaction and is obtained -- or it is polyether polyester which was made to carry out the ester interchange of polyol, dicarboxylic acid, and the polyvalent carboxylic acid (and/or, the acid anhydride) of three or more organic functions, and was obtained [ polyvalent carboxylic acid ] in dehydration, and the polyester and polyether polyol which be made to carry out a deglycol reaction and be obtained.

[0045] The polyether polyester (J) used for this invention is polyether polyester which carried out the ester interchange of the polyvalent carboxylic acid (and/or, the acid anhydride) of three or more organic functions to the polyether polyester which was made to carry out the ester interchange of polyol and the dicarboxylic acid, and was obtained [ dicarboxylic acid ] in dehydration and the polyester which is made to carry out a deglycol reaction and is obtained, and polyether polyol, it was made to react to it, and was obtained.

[0046] Furthermore, the polyether polyester (A) of these amounts of macromolecules, (H), (I), or (J) is the purpose which carries out macromolecule quantification of the molecular weight of the lactic-acid system copolymer obtained, and it is desirable that the molecular weight of polyether polyester (A), (H), (I), or (J) is 5,000-150,000 in 10,000-300,000, and number average molecular weight at weight average molecular weight.

[0047] Furthermore, it is more desirable that the polyether polyester (A) from a viewpoint of the solubility of polyether polyester and the ease of manufacture, (H), (I), or (J) is 10,000 to about 100,000 in 20,000-200,000, and number average molecular weight at weight average molecular weight.

[0048] Furthermore, as structure of polyether polyester (A), (H), (I), or (J), it is desirable from a viewpoint of solubility and biodegradability that it is aliphatic series polyether polyester. Furthermore, the aliphatic series polyether polyester with which polyether polyol consists more than of 20 mol % to

sum total mol % of polyether polyol and polyol excels and has the desirable thermal resistance of the lactic-acid system copolymer obtained.

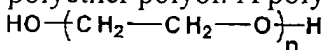
[0049] Moreover, the thermal resistance of the lactic-acid system copolymer of lactide (E) and polyether polyester ((A), (H), (I), (J)) comparatively obtained as it is 40 / 60 - 95/5 becomes high. furthermore, (A)/((A), (H), (I) or, and (J)) -- comparatively -- 50/50- the thermal resistance of the amount lactide system copolymer of macromolecules obtained as it is 80/20 becomes higher and is desirable.

[0050] As for the polyether polyester ((H), (I), (J)) used for this invention, the aliphatic series system polyether polyester with which a dicarboxylic acid component consists of aliphatic series dicarboxylic acid is mentioned. Moreover, although the aromatic series polyether polyester which consists of an aromatic series dicarboxylic acid component, or the aromatic series and aliphatic series system polyether polyester which consists of an aromatic series dicarboxylic acid component and an aliphatic series dicarboxylic acid component can also be used, when biodegradability is considered, aliphatic series polyether polyester is desirable.

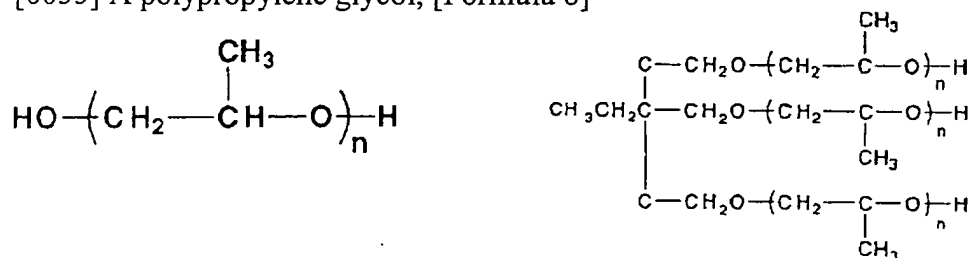
[0051] Although especially the aliphatic series dicarboxylic acid component in the polyether polyester used by this invention is not specified, it is desirable that it is the aliphatic series dicarboxylic acid (and those acid anhydrides), those methyl ester, and ethyl ester of the carbon atomic numbers 4-14. A succinic acid, an adipic acid, a sebacic acid, cyclohexane dicarboxylic acid and those methyl ester, ethyl ester, a succinic anhydride, a hexa hydro phthalic acid, etc. are desirable especially.

[0052] Although especially the aromatic series dicarboxylic acid component in the polyether polyester used for this invention is not specified, a phthalic acid, isophthalic acid, a terephthalic acid, 2, 6-naphthalene dicarboxylic acid, phthalic anhydride, dimer acid, etc. are specifically mentioned. In addition, alcohol with phthalic-acid, isophthalic acid, terephthalic-acid, 2, and 6-naphthalene dicarboxylic acid etc. and ester with polyol are mentioned.

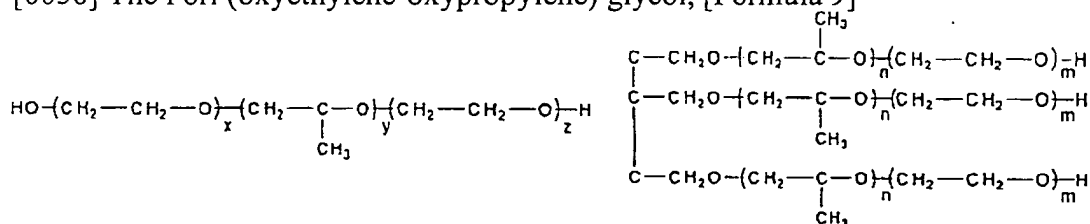
[0053] It is [0054], when a polyethylene glycol, a polypropylene glycol, the Pori (oxyethylene-oxypropylene) glycol that is the copolymer of ethyleneoxide and propylene oxide, Pori 1, 2-butylene glycol, Pori 1, 4-butylene glycol, and poly neopentyl glycol are desirable and consider biodegradability, versatility, etc. further especially, although a class will not be asked diol, triol, and more than it especially about the polyether polyol component in the polyether polyester used for this invention if it is polyether polyol. A polyethylene glycol, [Formula 7]



[0055] A polypropylene glycol, [Formula 8]



[0056] The Pori (oxyethylene-oxypropylene) glycol, [Formula 9]



Especially \*\* is desirable.

[0057] Moreover, as for the molecular weight of polyether polyol (A), it is desirable that it is 200-

200,000 in weight average molecular weight. Furthermore, it is more desirable that the viewpoint of biodegradability to polyether polyol (A) is 200 to about 10,000 in weight average molecular weight. [0058] Although a class will not be asked about the polyol component in polyether polyester especially if it is polyol, the polyol of 2-15 has a desirable carbon number especially, and it is specifically ethylene glycol, propylene glycol, a butylene glycol, pentane polyol, hexamethylene glycol, octane polyol, neopentyl glycol, cyclohexane dimethanol, a xylene glycol, and [0059]. a diethylene glycol, triethylene glycol, dipropylene glycol, and a jib -- hydrogenation bisphenol A, such as tongue diol, 3-hydroxy pivalyl pivalate, a glycerol, and pentaerythritol, is mentioned.

[0060] Although a class is not asked especially concerning the polyvalent-carboxylic-acid component of three or more organic functions, trimesic acid, propane tricarboxylic acid, trimellitic acid, trimellitic anhydride, pyromellitic acid, pyromellitic dianhydride, benzophenone tetracarboxylic acid, 1 and 3, 5-cyclohexane tricarboxylic acid, cyclohexane tetracarboxylic acid, a cyclohexane tetracarboxylic acid anhydride, etc. are mentioned especially.

[0061] Especially trimellitic anhydride and pyromellitic dianhydride are desirable. The above-mentioned polyvalent carboxylic acid can be mixed and used if needed. Especially, the amount of the polyvalent carboxylic acid used is explained below.

[0062] Polyether polyol (E), polyol (F), and dicarboxylic acid (G), In obtaining polyether polyester (H) dehydration and by carrying out a deglycol reaction, the polyvalent carboxylic acid (and/or, the acid anhydride) (D) of three or more organic functions The molecular weight of the lactic-acid system copolymer of the amount of macromolecules of the polyvalent carboxylic acid (D) of three or more organic functions comparatively obtained to a dicarboxylic acid (G) component as 0.001 - five-mol % is desirable and it is 0.01 - one-mol %, and flexibility become high.

[0063] And a deglycol reaction is carried out and it is obtained. polyol (F), dicarboxylic acid (G) and the polyvalent carboxylic acid of three or more organic functions, and/or its acid anhydride (D) -- dehydration -- with polyester In obtaining polyether polyester (I) by carrying out the ester exchange reaction of the polyether polyol (E) The molecular weight of the lactic-acid system copolymer of the amount of macromolecules of the polyvalent carboxylic acid (D) of three or more organic functions comparatively obtained to a dicarboxylic acid (G) component as 0.001 - five-mol % is desirable and it is especially 0.01 - one-mol %, and flexibility become high.

[0064] It is [0065] when obtaining polyether polyester (J) by carrying out the ester exchange reaction of the polyvalent carboxylic acid of three or more organic functions, and/or its acid anhydride (D) to polyol (F) and the polyether polyester which was made to carry out the ester interchange of the dicarboxylic acid (G), and was obtained [ dicarboxylic acid ] in dehydration and the polyester which is made to carry out a deglycol reaction and is obtained, and polyether polyol (E). 0.01 - 5 % of the weight is desirable to a component in the polyether polyester of the polyvalent carboxylic acid (D) of three or more organic functions which was made to carry out the ester interchange of the dicarboxylic acid (G) with polyol (F) comparatively, and was obtained in dehydration and the polyester which is made to carry out a deglycol reaction and is obtained, and polyether polyol (E), and the molecular weight of the lactic-acid system copolymer of the amount of macromolecules obtained as it is 0.1 - 1 % of the weight, and flexibility become high especially at it.

[0066] Branched chain is introduced by using the polyvalent carboxylic acid (D) of these three or more organic functions. The closure of the end of the effectiveness that molecular weight distribution are expanded to the increase list of the molecular weight of a copolymer, and the generated copolymer is carried out, and thermal stability increases, or a metal reacts to the carboxyl group of one or more functional groups of polyvalent carboxylic acid. When a polymer ionomer-izes, macromolecule quantification is easily possible and the lactic-acid system copolymer which can be fabricated on the film and sheet which have the outstanding physical properties can be manufactured.

[0067] If the lactic-acid system copolymer of the amount of macromolecules obtained by this invention is sheet-ized, from the sheet of high reinforcement to a flexible sheet can be obtained. Specifically, it is 500-50,000kg/cm<sup>2</sup> as hauling viscoelasticity. A sheet is obtained.

[0068] Macromolecule quantification can be further carried out by making it react with one or more

sorts of compounds chosen from the group which changes to the obtained lactic-acid system copolymer from hydroxy acid, carboxylic anhydrides, isocyanates, and lactone. If these examples are given, for example as hydroxy acid, a citric acid, a tartaric acid, a malic acid, etc. will be an acetic anhydride, a succinic anhydride, a propionic anhydride, phthalic anhydride, a maleic anhydride, a butyric anhydride, an isobutyric anhydride, trimellitic anhydride, pyromellitic dianhydride, etc. as carboxylic anhydrides. [0069] moreover -- isocyanate -- a kind -- \*\*\*\*\* -- phenyl -- isocyanate -- hexamethylene diisocyanate -- toluene 2,4-diisocyanate -- naphthalene - one -- five - diisocyanate -- a triphenylmethane color - four -- four -- ' -- four -- " - tri-isocyanate -- etc. -- lactone -- a kind -- \*\*\*\*\* -- beta - a caprolactone -- epsilon - a caprolactone -- beta - a butyrolactone -- delta - a valerolactone -- etc. -- mentioning -- having .

[0070] In order to make these compounds react to a lactic-acid system copolymer further, it mixes in the reactant after the polymerization reaction of a lactic-acid system copolymer is completed, and changes into the lactic-acid system copolymer which carried out stirring mixing in the state of short-time melting, or was obtained by the polymerization, and these may be added and melting mixing may be carried out. Moreover, after making it dissolve in a common solvent and making both lactic-acid system copolymer and these compounds heat and react to it, you may obtain by reprecipitation or degassing.

[0071] Especially, about hydroxy acid, carboxylic anhydrides, and lactone, the approach of adding these to the obtained lactic-acid system copolymer, and making it react to it under reduced pressure after melting mixing is desirable, and, thereby, reaction time becomes quick.

[0072] Generally 60 degrees C - 240 degrees C of temperature made to mix and react are 80 degrees C - 190 degrees C preferably. Moreover, on the occasion of a reaction, ester polymerization catalysts, such as N.N-dimethylaniline, tin octanoate, a dibutyl tin JIRAU rate, and isopropyl titanate, or an urethane catalyst can be used. Whenever [ reduced pressure / in the case of a reaction ] is 3 or less Torrs still more preferably 10 or less Torrs preferably 100 or less Torrs.

[0073] 0.001 % of the weight - 5% of the weight of a lactic-acid system copolymer is desirable still more desirable, and the amount of hydroxy acid, carboxylic anhydrides, isocyanates, and the lactone used is 0.001 % of the weight - 0.1 % of the weight. The used hydroxy acid, carboxylic anhydrides, isocyanates, and the lactone of this approach are desirable also at the point that it combines with the end group of a lactic-acid system copolymer, and the decomposition to the monomer of the polymer by heat can be prevented.

[0074] It is desirable to use a polymerization catalyst (C) for manufacture of this invention. As a polymerization catalyst (C) used by this invention, metals, such as the tin generally known also as the polymerization catalyst of cyclic ester and an ester interchange catalyst, zinc, lead, titanium, a bismuth, a zirconium, and germanium, and the derivative of those are mentioned. About especially these derivatives, an organic metal compound, a carbonate, an oxide, and a halogenide are desirable.

Specifically, tin octanoate, chlorination tin, a zinc chloride, zinc acetate, a lead oxide, lead carbonate, a titanium chloride, alkoxy titanium, the germanium dioxide, and the zirconium dioxide are suitable.

[0075] The amount of the polymerization catalyst (C) used has 0.005 - 0.2 desirable % of the weight to the sum total of lactide (B), polyether polyester ((A), (H), (I), or (J)), and/or the polyvalent carboxylic acid (D) of three or more organic functions, its rate of polymerization is fully quick, and especially in order to lessen coloring of the obtained lactide system polyether polyester, its 0.01 - 0.1 % of the weight is desirable.

[0076] Dehydration and in case it carries out a deglycol reaction and polyether polyester is manufactured, a catalyst can also be used for the polyvalent carboxylic acid (D), dicarboxylic acid, and polyol of three or more organic functions. As a catalyst used by this invention, as for each catalyst generally known as an esterification catalyst, metallic compounds organic [, such as tin, zinc, lead titanium, antimony, a cerium, germanium, cobalt manganese, iron, aluminum, magnesium, calcium, and strontium, / of at least a kind of metal ] or inorganic are mentioned usable.

[0077] For example, metaled alkoxide, an organic-acid salt, a chelate, oxide, etc. are used, and compounds, such as the organic compound of titanium, for example, titanic-acid alkyl ester, titanium oxy-acetylacetonate, and a titanium oxalate, are especially useful. As amount of the catalyst used, 0.001

- 0.5 % of the weight is desirable to the polyvalent carboxylic acid (D), the dicarboxylic acid, and the polyol sum total of three or more organic functions. A rate of polymerization is fully quick, and especially in order to lessen coloring of the obtained lactide system polyether polyester, 0.01 - 0.1 % of the weight is desirable.

[0078] Next, the concrete manufacture approach of this invention is explained. In case polyether polyester (A) and the polyvalent carboxylic acid (and/or, the acid anhydride) (D) of three or more organic functions are made to react, when it carries out under reduced pressure, a reaction goes on promptly and is desirable after melting mixing. the reaction which mixes lactide (E) with the macromolecule quantification polyether polyester obtained by the reaction, and performs a polymerization -- mixture -- warming -- melting is carried out or a polymerization catalyst (F) is added for a reactant after dilution mixing with a solvent.

[0079] polyether polyol (E), polyol (F), and dicarboxylic acid (G) -- and/or, the polymerization with the polyvalent carboxylic acid (and/or, the acid anhydride) (D) of three or more organic functions performs the dehydration and the deglycol reaction which are the synthetic approach of common polyester. the reaction which mixes lactide (B) with the obtained polyether polyester (H), and performs a polymerization -- mixture -- warming -- melting is carried out or a polymerization catalyst (F) is added for a reactant after dilution mixing with a solvent.

[0080] polyol (F) and dicarboxylic acid (G) -- and/or, the polymerization with the polyvalent carboxylic acid (and/or, the acid anhydride) (D) of three or more organic functions performs the dehydration and the deglycol reaction which are the synthetic approach of common polyester. Next, the ester exchange reaction of the polyether polyol (E) is carried out to the obtained polyester under reduced pressure after melting mixing. the reaction which mixes lactide (B) with the obtained polyether polyester (I), and performs a polymerization -- mixture -- warming -- melting is carried out or a polymerization catalyst (F) is added for a reactant after dilution mixing with a solvent.

[0081] Polyol (F) and the polymerization with dicarboxylic acid (G) perform the dehydration and the deglycol reaction which are the synthetic approach of common polyester. Next, the ester exchange reaction of the polyether polyol (E) is carried out to the obtained polyester under reduced pressure after melting mixing. Next, the ester exchange reaction of the obtained polyether polyester and the polyvalent carboxylic acid (and/or, the acid anhydride) of three or more organic functions is carried out under reduced pressure after melting mixing.

[0082] the reaction which mixes lactide (B) with the obtained polyether polyester (J), and performs a polymerization -- mixture -- warming -- melting is carried out or a polymerization catalyst (F) is added for a reactant after dilution mixing with a solvent. When the dehydration and the deglycol reaction which are the synthetic approach of common polyester here are described briefly, as for dehydration, it is desirable to carry out under an inert gas ambient atmosphere for 150-250 degrees C and 3 to 16 hours.

[0083] The deglycol reaction is gradually carried out to reduced pressure, and, finally it is desirable under reduced pressure of 5 or less Torrs to carry out at 170-260 degrees C for 2 to 16 hours. thus, the polyether polyester ((A), (H), (I) or, and (J)) and lactide (E) which were obtained -- warming -- when carrying out melting or adding a polymerization catalyst (F) after mixing by solvent addition, a polymerization system is made to homogeneity as it is more than the melting point of lactide, a quick rate of polymerization is obtained, and polymerization temperature is desirable.

[0084] The polymerization temperature in a non-solvent system is more than the melting point of lactide, is desirable on the balance of a polymerization, and can prevent coloring of the lactide system polyether polyester accompanying a decomposition reaction. [ of the temperature of 185 degrees C or less ] The melting point of lactide is near 100 degree C, and can prevent the temperature of 185 degrees C or less of 100 degrees C or more, the fall of the molecular weight of the lactide system polyether polyester accompanying [ 145-180 degrees C is desirable on the balance of a polymerization still more preferably, and ] a decomposition reaction, and coloring.

[0085] moreover, the thing for which all reactions are performed under the dry inert gas ambient atmosphere in order to prevent decomposition of lactide, and coloring -- it is desirable. It carries out in

the state of bubbling under nitrogen and an argon gas ambient atmosphere especially. It is desirable that the polyether polyol and polyether polyester it is incomparable in a raw material also remove moisture to coincidence, and dry it by reduced pressure drying etc.

[0086] Moreover, since lactide can be dissolved in a solvent, a polymerization can be carried out using a solvent and benzene, toluene, ethylbenzene, a xylene, a cyclohexanone, a methyl ethyl ketone, isopropyl ether, etc. are mentioned as an example of a solvent, for example.

[0087] The thing which used the usual polymerization iron pot and to do for batch manufacture is also possible for the lactic-acid system copolymer of the amount of giant molecules of this invention. Furthermore, improvement in product quality, yield, and productivity can be aimed at by the continuation manufacture approach. As the continuation manufacture approach, the stirring type reactor connected with two or more serials, a static mixer, and/or a horizontal-type reactor are used for all or a part of the reaction processes.

[0088] Use of the stirring type reaction vessel which changed the stirring approach with advance of a reaction and was connected with the serial of two or more tubs in which efficient stirring and temperature control are possible as a stirring type reactor connected with two or more serials is desirable.

[0089] The static mixer which says here is static mixing equipment which the amount of moving part is not to the mixed equipment which has an agitator, namely, does not have an agitator, flow divides, and a flow direction converts or reverses by the mixing element the amount of [ which was fixed in tubing ] moving part is not specifically, and the mixed equipment which mixes a solution says by repeating division, conversion, and reversal for flow in a lengthwise direction and a longitudinal direction.

Moreover, of course, an extruder [ like one shaft used for reactive processing or a biaxial extruder ] whose horizontal-type reactor is says the thing of the biaxial reactor through which it can circulate.

[0090] Even if a solvent etc. is used for these continuation manufacture approach, it can carry out a polymerization. Although the lactic-acid system copolymer of the obtained amount of macromolecules has high melt viscosity the top where the melting point is high and it is hard to carry out a polymerization by this, it falls, stirring becomes easy and the viscosity of a polymerization system becomes easy to perform a polymerization by adding a solvent.

[0091] the polymerization equipment which the knockout pressure of a polymerization solution fell and had the interior equipment for heat carriers, and a baffle aiming at stirring for the purpose of the temperature control when continuation polymerization equipment equipped with especially the static mixer was used -- equipment -- casual dress -- it does-izing and is effective. Since stirring is easy, a temperature control is easy, temperature is homogeneous in polymerization equipment, and the lactic-acid system copolymer of the amount of macromolecules with less coloring etc. is obtained.

[0092] Moreover, it is desirable to devolatilize under reduced pressure in order to remove the matter with the lactide, solvent, and odor which remained at the polymerization anaphase. The amount of residual lactide can be decreased according to this devolatilization process, and the preservation stability of the lactic-acid system copolymer of the obtained amount of macromolecules can be increased remarkably.

[0093] Residual lactide becomes hydrolysis by adhesion of moisture etc., or the cause of the welding by heat and is not desirable when the amount lactide system polyether polyester of macromolecules is used as a sheet film. Moreover, it disperses [ by sublimation ] from the film sheet produced commercially and is not desirable. As for the amount of residual lactide in the lactic-acid system copolymer of this invention, for this reason, it is desirable to carry out to 2 or less % of the weight. It carries out to 1 or less % of the weight still more preferably.

[0094] As the approach of concrete devolatilization, one shaft or a biaxial extruder, a thin film distillation machine, a pot mold decompression device, etc. perform. As conditions for devolatilization, the method of performing ejection is desirable under reduced pressure after a polymerization, heating. In order not to reduce the molecular weight of a lactic-acid system copolymer, as for devolatilization conditions, it is desirable that whenever [ 100-230 degrees-C and reduced pressure ] performs devolatilization time amount, and temperature performs it by 0.1 - 200Torr for 10 seconds to 10



minutes.

[0095] There is a method of performing ejection pelletizing or grinding a lactic-acid system copolymer, and heating it under reduced pressure after polymerization termination, as the other devolatilization approaches. 0.1 - 50Torr of temperature is [ devolatilization time amount / whenever / 60-200-degree-C and reduced pressure ] desirable in order not to reduce the molecular weight of a lactic-acid system copolymer in this case, either for 2 to 400 minutes.

[0096] In case the copolymer of this invention is manufactured, cyclic ester other than lactide (E) can be added further, and the lactic-acid system copolymer of the amount of macromolecules can also be made. Lactone can be added one to 20% of the weight especially for the purpose of elasticity-izing. About the cyclic ester added in addition to lactide, although there is especially no constraint, the annular dimerization object of hydroxy acids, such as glycolide, intramolecular lactide especially beta-butyrolactone, epsilon-caprolactone, gamma-valerolactone, g undecalactone, etc. are specifically mentioned. If the amount of lactone increases, a glass transition point and the melting point will become low, and flexibility will become high.

[0097] In case lactic-acid system copolymerization of this invention is formed on a sheet film etc., mixing addition of the organic system bulking agents, such as inorganic system bulking agents, such as a general filler, for example, talc, a calcium carbonate, a silica, clay, the diatom earth, and a pearlite, or wood flour, may be carried out. Moreover, stabilizers, such as ultraviolet ray absorbents, such as 2,6-di-tert-butyl-4-methylphenol (BHT), an anti-oxidant like a butylhydroxyanisol (BHA), salicylic acid derivatives, a benzophenone system, and a benzotriazol system, and phosphoric ester, and a carbodiimide, can be used, and the thermal stability at the time of shaping can be raised.

[0098] Although the lactic-acid system copolymer of this invention has sufficient plasticity independently and has a good moldability, when planning still higher fabrication nature and flexibility, it may add plasticizers, such as dioctyl adipate, a dioctyl sebacate, trioctyl trimellitate, a diethyl phthalate, a dioctyl phthalate, a polypropylene-glycol adipic acid, and adipic-acid butanediol.

[0099] Moreover, even if a coloring agent like surface active agents, such as ion systems, such as non-ion systems, such as lubricant, such as metallic soap, such as zinc stearate, magnesium stearate, and stearic acid calcium, mineral oil, a liquid paraffin, and ethylene screw stearyl AMAIDO, a glycerine fatty acid ester, and cane-sugar fatty acid ester, and an alkyl sulfonate, titanium oxide, and carbon black etc. adds, it does not interfere.

[0100] moreover, addition of organic system foaming agents, such as inorganic system foaming agents, such as sodium bicarbonate and ammonium bicarbonate, an AZOJI carvone amide, azobisisobutyronitril, and sulfonylhydrazide, etc. -- or -- or it infiltrates foaming agents, such as a pentane, butane, and Freon, into the lactic-acid system copolymer of this invention in advance -- an extrusion process -- on the way -- it can also consider as foam by coming out and supplying directly in an extruder. Moreover, multilayering with paper, aluminum foil, or other resolvability polymer films is also possible by extrusion lamination, dry lamination, or co-extrusion.

[0101] The lactic-acid system polyether polyester copolymer obtained by this invention has good biodegradability, and even if it is discarded after being used for general-purpose resin, wrapping, etc., or discarded from a production process, it is useful to loss in quantity of trash. When abandoned all over soil and seawater, decomposition by hydrolysis, a microorganism, etc. is received and the reinforcement as resin deteriorates in several months, and by the time it does not maintain an appearance, it can decompose. Moreover, if compost is used, biodegradation will be carried out by the time it does not stop the original form further for a short period of time.

[0102]

[Example] This invention is not limited by these examples although an example and the example of a comparison explain this invention still more concretely below. In addition, especially, the polyether polyol in an example used the diol mold altogether, as long as it was unstated.

[0103] In addition, the determination of molecular weight was measured by the comparison with a polystyrene correlation sample with the GPC measuring device (Following GPC, abbreviation, the column temperature of 40 degrees C, tetrahydrofuran solvent). The differential-scanning-calorie meter



(Following DSC and abbreviation) performed a glass transition point and the melting point. The temperature up of the Measuring condition was carried out from 20 degrees C to 220 degrees C by 10 degrees C / min.

[0104] Moreover, the Measuring condition of a tension test was performed by 50mm of initial sample length, and crosshead rate 40 mm/min. The test piece cut and measured the film of 250-micrometer thickness created under 160-180 degrees C, 200kg/cm<sup>2</sup>, and the conditions for 2 minutes with the hotpress machine in width-of-face [ of 15mm ] x die length of 80mm.

[0105] In addition, the above-mentioned device used the device described below.

GPC:TOSOH HLC-8020 (TOSOH CORP. make)

DSC:DSC 200 (the SEIKO electronic industry incorporated company make)

Tension test: Tensilon (Oriental energy machine incorporated company make)

[0106] [Example 1] Heating stirring was carried out, teaching polypropylene-glycol 45g of number average molecular weight 400, ethylene glycol 4.1g, and 25g of sebacic acids to 300ml 4 opening flask which attached an agitator, a rectifier, and gas installation tubing, and carrying out a temperature up to it in 150 degrees C to 10 degrees C /under nitrogen-gas-atmosphere mind in an hour.

[0107] The temperature up was carried out to 220 degrees C, distilling off the water to generate, after checking that the distillate of water has stopped, titanium isopropyl titanate 0.01g was added and the deglycol reaction was performed at 220 degrees C under reduced pressure of 0.5Torr(s) for 3 hours. it is [ polyester / which was obtained / polyether ] \*\*\*\*\* in number average molecular weight 18,000 and weight average molecular weight 33,000 -- it was liquefied.

[0108] It taugt 200ml separable flask, and under nitrogen-gas-atmosphere mind, it fused, obtained polyether polyester 30g, L-lactide 68g, D-lactide 2g, and 175-degree C toluene 10ml were mixed for 0.25 hours, and 0.03g of tin octanoate was added. The generated copolymer was taken out after the 3-hour reaction.

[0109] The obtained lactic-acid system polyether polyester was transparent, and was number average molecular weight 55,000 and weight average molecular weight 93,000 from GPC. The glass transition point was observed by about 50 degrees C, and the melting point was about 156 degrees C. Moreover, fracture distortion was [ 390 kgf/cm<sup>2</sup> and the initial elastic modulus of of breaking strength ] 13,000 kgf/cm<sup>2</sup> 10%.

[0110] [Example 2] Heating stirring was carried out, teaching ethylene glycol 21g and 50g of sebacic acids to 300ml 4 opening flask which attached an agitator, a rectifier, and gas installation tubing, and carrying out a temperature up to it in 150 degrees C to 10 degrees C /under nitrogen-gas-atmosphere mind in an hour. The temperature up was carried out to 220 degrees C, distilling off the water to generate, after checking that the distillate of water has stopped, titanium isopropyl titanate 0.01g was added and the deglycol reaction was performed at 220 degrees C under reduced pressure of 0.5Torr(s) for 3 hours. The obtained polyester (A-1) was the white solid-state of number average molecular weight 20,000 and weight average molecular weight 35,000.

[0111] Obtained polyester (A-1) 13g and polypropylene-glycol 60g of number average molecular weight 3,000 were taught to 200ml 4 opening flask which attached an agitator, a rectifier, and gas installation tubing, and the deglycol reaction was performed at 220 degrees C under reduced pressure of 0.5Torr for 3 hours. it is [ polyester / (A-2) / which was obtained / polyether ] \*\*\*\*\* in number average molecular weight 21,000 and weight average molecular weight 36,000 -- it was liquefied.

[0112] It taught 200ml separable flask, and under nitrogen-gas-atmosphere mind, it fused, obtained polyester 10g, L-lactide 85g, D-lactide 5g, and 175-degree C toluene 10ml were mixed for 0.25 hours, and 0.03g of tin octanoate was added. The generated copolymer was taken out after the 3-hour reaction.

[0113] The obtained lactic-acid system polyether polyester was transparent, and was number average molecular weight 87,000 and weight average molecular weight 150,000 from GPC. The glass transition point was observed by about 53 degrees C, and the melting point was about 158 degrees C. Moreover, fracture distortion was [ 490 kgf/cm<sup>2</sup> and the initial elastic modulus of of breaking strength ] 21,000 kgf/cm<sup>2</sup> 6%.

[0114] [Example 3] It taught 200ml separable flask, and under nitrogen-gas-atmosphere mind, it fused, polyether polyester (A-2) 30g obtained in the example 2, L-lactide 68g, D-lactide 2g, and 175-degree C toluene 10ml were mixed for 0.25 hours, and 0.03g of tin octanoate was added.

[0115] The generated copolymer was taken out after the 3-hour reaction. The obtained lactic-acid system polyether polyester was transparent, and was number average molecular weight 72,000 and weight average molecular weight 120,000 from GPC. Observed by about 53 degrees C although glass transition points were few, the melting point was about 158 degrees C. Moreover, \*\*\*\* fracture distortion was [ 420 kgf/cm<sup>2</sup> and the initial elastic modulus of \*\*\*\* of \*\*\*\* breaking strength ] 19,000 kgf/cm<sup>2</sup> 11%.

[0116] [Example 4] It taught 200ml separable flask, and under nitrogen-gas-atmosphere mind, it fused, polyether polyester (A-2) 50g obtained in the example 2, L-lactide 48g, D-lactide 2g, and 175-degree C toluene 10ml were mixed for 0.25 hours, and 0.03g of tin octanoate was added. The generated copolymer was taken out after the 3-hour reaction.

[0117] The obtained lactic-acid system polyether polyester was transparent, and was number average molecular weight 52,000 and weight average molecular weight 71,000 from GPC. Observed by about 53 degrees C although glass transition points were few, the melting point was about 153 degrees C. moreover, \*\*\*\* fracture distortion -- 200% and \*\*\*\* breaking strength -- 350 kgf/cm<sup>2</sup> and the initial elastic modulus of \*\*\*\* -- 6,200 kgf/cm<sup>2</sup> it was .

[0118] [Example 5] Heating stirring was carried out, teaching polypropylene-glycol 90g of number average molecular weight 3,000, ethylene glycol 2.0g, and 8.7g of sebacic acids to 300ml 4 opening flask which attached an agitator, a rectifier, and gas installation tubing, and carrying out a temperature up to it in 150 degrees C to 10 degrees C /under nitrogen-gas-atmosphere mind in an hour. Distilling off the water to generate, after checking that carried out the temperature up to 220 degrees C, and the distillate of water has stopped, titanium isopropyl titanate 0.01g was added and the deglycol reaction was performed at 220 degrees C under reduced pressure of 0.5Torr(s) for 3 hours.

[0119] it is [ polyester / which was obtained / polyether ] \*\*\*\*\* in number average molecular weight 19,000 and weight average molecular weight 32,000 -- it was liquefied. It taught 200ml separable flask, and under nitrogen-gas-atmosphere mind, it fused, obtained polyether polyester 30g, L-lactide 68g, D-lactide 2g, and 175-degree C toluene 10ml were mixed for 0.25 hours, and 0.03g of tin octanoate was added.

[0120] The generated copolymer was taken out after the 3-hour reaction. The obtained lactic-acid system polyether polyester was transparent, and was number average molecular weight 45,000 and weight average molecular weight 88,000 from GPC. The glass transition point was observed by about 50 degrees C, and the melting point was about 156 degrees C. Moreover, \*\*\*\* fracture distortion was [ 360 kgf/cm<sup>2</sup> and the initial elastic modulus of \*\*\*\* of \*\*\*\* breaking strength ] 18,000 kgf/cm<sup>2</sup> 10%.

[0121] [Example 6] Heating stirring was carried out, teaching propylene glycol 26g and 50g of sebacic acids to 300ml 4 opening flask which attached an agitator, a rectifier, and gas installation tubing, and carrying out a temperature up to it in 150 degrees C to 10 degrees C /under nitrogen-gas-atmosphere mind in an hour. The temperature up was carried out to 220 degrees C, distilling off the water to generate, after checking that the distillate of water has stopped, titanium isopropyl titanate 0.01g was added and the deglycol reaction was performed at 220 degrees C under reduced pressure of 0.5Torr(s) for 3 hours.

[0122] it is [ polyester / which was obtained ] \*\*\*\*\* in number average molecular weight 17,000 and weight average molecular weight 28,000 -- it was liquefied. Obtained polyester 14g and polypropylene-glycol 60g of number average molecular weight 3,000 were taught to 200ml 4 opening flask which attached an agitator, a rectifier, and gas installation tubing, and the deglycol reaction was performed at 220 degrees C under reduced pressure of 0.5Torr for 3 hours. it is [ polyester / which was obtained / polyether ] \*\*\*\*\* in number average molecular weight 20,000 and weight average molecular weight 35,000 -- it was liquefied.

[0123] It taught 200ml separable flask, and under nitrogen-gas-atmosphere mind, it fused, obtained polyether polyester 30g, L-lactide 68g, D-lactide 2g, and 175-degree C toluene 10ml were mixed for

0.25 hours, and 0.03g of tin octanoate was added. The generated copolymer was taken out after the 3-hour reaction.

[0124] The obtained lactic-acid system polyether polyester was transparent, and was number average molecular weight 61,000 and weight average molecular weight 120,000 from GPC. The glass transition point was observed by about 53 degrees C, and the melting point turned out to be about 152 degrees C. moreover, \*\*\*\* fracture distortion -- 15% and \*\*\*\* breaking strength -- 330 kgf/cm<sup>2</sup> and the initial elastic modulus of \*\*\*\* -- 12,000 kgf/cm<sup>2</sup> it was .

[0125] [Example 7] Heating stirring was carried out, teaching polyethylene-glycol 110g of number average molecular weight 2,000, ethylene glycol 3.2g, and 16g of sebacic acids to 300ml 4 opening flask which attached an agitator, a rectifier, and gas installation tubing, and carrying out a temperature up to it in 150 degrees C to 10 degrees C /under nitrogen-gas-atmosphere mind in an hour.

[0126] The temperature up was carried out to 220 degrees C, distilling off the water to generate, after checking that the distillate of water has stopped, titanium isopropyl titanate 0.01g was added and the deglycol reaction was performed at 220 degrees C under reduced pressure of 0.5Torr(s) for 3 hours. The obtained polyether polyester was the solid-state of number average molecular weight 18,000 and weight average molecular weight 33,000.

[0127] It taught 200ml separable flask, and under nitrogen-gas-atmosphere mind, it fused, obtained polyether polyester 30g, L-lactide 68g, D-lactide 2g, and 175-degree C toluene 10ml were mixed for 0.25 hours, and 0.03g of tin octanoate was added. The generated copolymer was taken out after the 3-hour reaction.

[0128] The obtained lactic-acid system polyether polyester was transparent, and was number average molecular weight 45,000 and weight average molecular weight 88,000 from GPC. The glass transition point was observed by about 50 degrees C, and the melting point was about 156 degrees C. moreover, \*\*\*\* fracture distortion -- 120% and \*\*\*\* breaking strength -- 200 kgf/cm2 and the initial elastic modulus of \*\*\*\* -- 7,100 kgf/cm2 it was .

[0129] [Example 8] Heating stirring was carried out, teaching polyethylene-glycol 140g of number average molecular weight 20,000, ethylene glycol 1.0g, and 2.0g of sebacic acids to 300ml 4 opening flask which attached an agitator, a rectifier, and gas installation tubing, and carrying out a temperature up to it in 150 degrees C to 10 degrees C /under nitrogen-gas-atmosphere mind in an hour.

[0130] The temperature up was carried out to 220 degrees C, distilling off the water to generate, after checking that the distillate of water has stopped, titanium isopropyl titanate 0.01g was added and the deglycol reaction was performed at 220 degrees C under reduced pressure of 0.5Torr(s) for 3 hours. The obtained polyether polyester was the solid-state of number average molecular weight 42,000 and weight average molecular weight 58,000.

[0131] It taught 200ml separable flask, and under nitrogen-gas-atmosphere mind, it fused, obtained polyether polyester 30g, L-lactide 68g, D-lactide 2g, and 175-degree C toluene 10ml were mixed for 0.25 hours, and 0.03g of tin octanoate was added. The generated copolymer was taken out after the 3-hour reaction.

[0132] The obtained lactic-acid system polyether polyester was transparent, and was number average molecular weight 72,000 and weight average molecular weight 130,000 from GPC. The glass transition point was observed by about 50 degrees C, and the melting point was about 152 degrees C. moreover, \*\*\*\* fracture distortion -- 94% and \*\*\*\* breaking strength -- 210 kgf/cm<sup>2</sup> and the initial elastic modulus of \*\*\*\* -- 6,800 kgf/cm<sup>2</sup> it was .

[0133] [Example 9] Heating stirring was carried out, teaching ethylene glycol 57g and 100g of adipic acids to 300ml 4 opening flask which attached an agitator, a rectifier, and gas installation tubing, and carrying out a temperature up to it in 150 degrees C to 10 degrees C /under nitrogen-gas-atmosphere mind in an hour. The temperature up was carried out to 200 degrees C, distilling off the water to generate, and the deglycol reaction was performed for 30 minutes at 200 degrees C under reduced pressure of 3Torr after checking that the distillate of water has stopped.

[0134] Polypropylene-glycol 714g of number average molecular weight 3,000 and titanium isopropyl titanate 0.01g were added to this, and the deglycol reaction was performed to it at 200 degrees C under

reduced pressure of 0.5Torr(s) for 3 hours. it is [ polyester / (A-3) / which was obtained / polyether ]  
 \*\*\*\*\* in number average molecular weight 25,000 and weight average molecular weight 40,000 -  
 - it was liquefied.

[0135] It taught 200ml separable flask, and under nitrogen-gas-atmosphere mind, it fused, obtained polyether polyester 30g, L-lactide 68g, D-lactide 2g, and 175-degree C toluene 10ml were mixed for 0.25 hours, and 0.03g of tin octanoate was added. The generated copolymer was taken out after the 3-hour reaction.

[0136] The obtained lactic-acid system polyether polyester was transparent, and was number average molecular weight 610,000 and weight average molecular weight 11,000 from GPC. The glass transition point was observed by about 50 degrees C, and the melting point was about 152 degrees C. Moreover, \*\*\*\* fracture distortion was [ 280 kgf/cm<sup>2</sup> and the initial elastic modulus of \*\*\*\* of \*\*\*\* breaking strength ] 10,000 kgf/cm<sup>2</sup> 27%.

[0137] [Example 10] Heating stirring was carried out, teaching ethylene glycol 71g and 100g of amber acids to 3l. 4 opening flask which attached an agitator, a rectifier, and gas installation tubing, and carrying out a temperature up to it in 150 degrees C to 10 degrees C /under nitrogen-gas-atmosphere mind in an hour. The temperature up was carried out to 200 degrees C, distilling off the water to generate, and the deglycol reaction was performed at 200 degrees C under reduced pressure of 3Torr after checking that the distillate of water has stopped for 30 minutes.

[0138] Polypropylene-glycol 890g of number average molecular weight 3,000 and titanium isopropyl titanate 0.01g were added to this, and the deglycol reaction was performed to it at 200 degrees C under reduced pressure of 0.5Torr(s) for 3 hours. it is [ polyester / which was obtained ] \*\*\*\*\* in number average molecular weight 22,000 and weight average molecular weight 39,000 -- it was liquefied.

[0139] It taught 200ml separable flask, and under nitrogen-gas-atmosphere mind, it fused, obtained polyether polyester 30g, L-lactide 68g, D-lactide 2g, and 175-degree C toluene 10ml were mixed for 0.25 hours, and 0.03g of tin octanoate was added. The generated copolymer was taken out after the 3-hour reaction.

[0140] The obtained lactic-acid system polyether polyester was transparent, and was number average molecular weight 720,000 and weight average molecular weight 12,000 from GPC. The glass transition point was observed by about 50 degrees C, and the melting point turned out to be about 152 degrees C. Moreover, \*\*\*\* fracture distortion was [ 270 kgf/cm<sup>2</sup> and the initial elastic modulus of \*\*\*\* of \*\*\*\* breaking strength ] 10,000 kgf/cm<sup>2</sup> 29%.

[0141] [Example 11] Polyether polyester (A-3) 30g obtained in the example 9 and 0.18g (it abbreviates to PMDA henceforth) of pyromellitic dianhydride were taught to 300ml 4 opening flask which attached an agitator, a rectifier, and gas installation tubing, and the deglycol reaction was performed at 200 degrees C under reduced pressure of 0.5Torr for 3 hours. it is [ polyester / which was obtained ] \*\*\*\*\* in number average molecular weight 33,000 and weight average molecular weight 90,000 -  
 - it was liquefied.

[0142] It taught 200ml separable flask, and under nitrogen-gas-atmosphere mind, it fused, obtained polyether polyester 30g, L-lactide 68g, D-lactide 2g, and 175-degree C toluene 10ml were mixed for 0.25 hours, and 0.03g of tin octanoate was added. The generated copolymer was taken out after the 3-hour reaction.

[0143] The obtained lactic-acid system polyether polyester was transparent, number average molecular weight 65,000, weight average molecular weight 13,000, and a glass transition point were observed by about 53 degrees C from GPC, and the melting point was about 150 degrees C. Moreover, \*\*\*\* fracture distortion was [ 220 kgf/cm<sup>2</sup> and the initial elastic modulus of \*\*\*\* of \*\*\*\* breaking strength ] 9,100 kgf/cm<sup>2</sup> 43%.

[0144] [Example 12] Heating stirring was carried out, teaching ethylene glycol 57g, 100g of adipic acids, and 0.6g of pyromellitic dianhydride to 300ml 4 opening flask which attached an agitator, a rectifier, and gas installation tubing, and carrying out a temperature up to it in 150 degrees C to 10 degrees C /under nitrogen-gas-atmosphere mind in an hour. The temperature up was carried out to 200

degrees C, distilling off the water to generate, and the deglycol reaction was performed at 200 degrees C under reduced pressure of 3Torr after checking that the distillate of water has stopped for 30 minutes.

[0145] Polypropylene-glycol 719g of number average molecular weight 3,000 and titanium isopropyl titanate 0.01g were added to this, and the deglycol reaction was performed to it at 200 degrees C under reduced pressure of 0.5Torr(s) for 3 hours. it is [ polyester / which was obtained / polyether ]

\*\*\*\*\* in number average molecular weight 25,000 and weight average molecular weight 40,000 - it was liquefied.

[0146] It taught 200ml separable flask, and under nitrogen-gas-atmosphere mind, it fused, obtained polyether polyester 30g, L-lactide 68g, D-lactide 2g, and 175-degree C toluene 10ml were mixed for 0.25 hours, and 0.03g of tin octanoate was added. The generated copolymer was taken out after the 3-hour reaction. The obtained lactic-acid system polyether polyester was transparent, and was number average molecular weight 51,000 and weight average molecular weight 110,000 from GPC. The glass transition point was observed by about 50 degrees C, and the melting point was about 152 degrees C. Moreover, \*\*\*\* fracture distortion was [ 330 kgf/cm<sup>2</sup> and the initial elastic modulus of \*\*\*\* of \*\*\*\* breaking strength ] 10,000 kgf/cm<sup>2</sup> 31%.

[0147] [Example 13] 80g (PE[ by Sanyo Chemical Industries, Ltd. ]- 75) of block copolymers of the polyethylene glycol and polypropylene glycol of polyester (A-1) 13g and number average molecular weight 4,025 obtained in the example 2 was taught to 200ml 4 opening flask which attached an agitator, a rectifier, and gas installation tubing, and the deglycol reaction was performed at 220 degrees C under reduced pressure of 0.5Torr for 3 hours. it is [ polyester / which was obtained / polyether ] \*\*\*\*\* in number average molecular weight 25,000 and weight average molecular weight 44,000 -- it was liquefied.

[0148] It taught 200ml separable flask, and under nitrogen-gas-atmosphere mind, it fused, obtained polyether polyester 30g, L-lactide 68g, D-lactide 2g, and 175-degree C toluene 10ml were mixed for 0.25 hours, and 0.03g of tin octanoate was added. The generated copolymer was taken out after the 3-hour reaction.

[0149] The obtained lactic-acid system polyether polyester was transparent, number average molecular weight 57,000, weight average molecular weight 83,000, and a glass transition point were observed by about 50 degrees C from GPC, and the melting point was about 152 degrees C. moreover, \*\*\*\* fracture distortion -- 32% and \*\*\*\* breaking strength -- 310 kgf/cm<sup>2</sup> and the initial elastic modulus of \*\*\*\* -- 10,000 kgf/cm<sup>2</sup> it was .

[0150] [Example 14] Polyester (A-1) 11g obtained in the example 2, 90g (PE[ by Sanyo Chemical Industries, Ltd. ]- 75) of block copolymers of the polyethylene glycol and polypropylene glycol of number average molecular weight 4,025, 10g (GL[ by Sanyo Chemical Industries, Ltd. ]- 3000) of triol mold block copolymers of the polyethylene glycol and polypropylene glycol of number average molecular weight 3,000 An agitator, It taught 200ml 4 opening flask which attached a rectifier and gas installation tubing, and the deglycol reaction was performed at 220 degrees C under reduced pressure of 0.5Torr for 3 hours.

[0151] it is [ polyester / which was obtained / polyether ] \*\*\*\*\* in number average molecular weight 31,000 and weight average molecular weight 59,000 -- it was liquefied. It taught 200ml separable flask, and under nitrogen-gas-atmosphere mind, it fused, obtained polyether polyester 30g, L-lactide 68g, D-lactide 2g, and 175-degree C toluene 10ml were mixed for 0.25 hours, and 0.03g of tin octanoate was added. The generated copolymer was taken out after the 3-hour reaction.

[0152] The obtained lactic-acid system polyether polyester was transparent, and was number average molecular weight 79,000 and weight average molecular weight 130,000 from GPC. The glass transition point was observed by about 51 degrees C, and the melting point was about 152 degrees C. Moreover, \*\*\*\* fracture distortion was [ 290 kgf/cm<sup>2</sup> and the initial elastic modulus of \*\*\*\* of \*\*\*\* breaking strength ] 10,000 kgf/cm<sup>2</sup> 40%.

[0153] The hotpress was used for the lactic-acid system copolymerization polyether polyester obtained in the example 3, and the sheet (thickness 10cmx10cm and 100 micrometers) was created. The weight average molecular weight of the sheet obtained under 165 degrees C, 200kg/cm<sup>2</sup>, and the press

conditions for 2 minutes was 117,000. This sheet was laid underground into soil and the biodegradation test was tried. A result is shown in Table 1.

[0154]

[Table 1]

浸漬日数	重量平均分子量	外観
0日	117,000	剛直 透明
30日	52,000	剛直 半透明
100日	15,000	脆い 白色
200日	6,200	脆い 白色

[0155] [Example 1 of a comparison] Toluene 15ml was added to L-lactide 100g, under nitrogen-gas-atmosphere mind, the dissolution and mixing of were done, 0.03g of tin octanoate was added as a ring-opening-polymerization catalyst, and 175 degrees C of polymerizations were performed for 3 hours for 0.25 hours. Toluene was removed under reduced pressure after the reaction. Generated L-poly(lactic acid) was transparent and colorless resin of weight average molecular weight 273,000 and number average molecular weight 140,000. The glass transition point of this was about 57 degrees C, and the melting point was about 158 degrees C. moreover, \*\*\*\* fracture distortion -- 3% and \*\*\*\* breaking strength -- 500 kgf/cm<sup>2</sup> and the initial elastic modulus of \*\*\*\* -- 16,000 kgf/cm<sup>2</sup> it was .

[0156] [the example 2 of a comparison] -- L-lactide 70g -- poly caprolactone (product made from UCC "tone") 30g, and toluene 15g -- in addition, under nitrogen-gas-atmosphere mind, the dissolution and mixing of were done, 175 degrees C of 0.03g of tin octanoate were added as a ring-opening-polymerization catalyst for 0.25 hours, and the polymerization was performed for 3 hours. Toluene was removed under reduced pressure after the reaction. The generated lactide system copolymerization polyether polyester was resin of the white of weight average molecular weight 223,000 and number average molecular weight 110,000. For about 47 degrees C and the melting point, about 149 degrees C and \*\*\*\* fracture distortion were [ the glass transition point of this / 250 kgf/cm<sup>2</sup> and the initial elastic modulus of \*\*\*\* of \*\*\*\* breaking strength ] 13,000 kgf/cm<sup>2</sup> 30%.

[0157] The measurement result about the lactic-acid system copolymer obtained in the example and the example of a comparison is indicated from Table 2 to a front table. In addition, the abbreviation of front Naka shows the following contents.

[0158] sebacic-acid: -- SeA adipic-acid: -- AA succinic-acid: -- SA pyromellitic dianhydride :P MDA

[0159] Ethylene glycol: EG propylene glycol :P G polyethylene glycol :P EG polypropylene glycol :P

The triol mold block copolymer of PGPEG, block-copolymer:EbPPEG which is PPG, and PPG: tEbP

[0160]

[Table 2]

項 目		実施例 1	実施例 2	実施例 3	実施例 4
L-LD (重量%)		68	85	68	48
D-LD (重量%)		2	5	2	2
ポリ エ ー テ ル エ ス テ ル	組成	SeA/EG/PPG	SeA/EG/PPG	SeA/EG/PPG	SeA/EG/PPG
	組成比(モル%)	100/10/90	100/75/35	100/75/35	100/75/35
	ポリエーテルリオル のMw	400	3000	3000	3000
	量 (重量%)	30	10	30	50
	Mw $\times 10^{-3}$	33	35	35	35
Mw $\times 10^{-3}$		93	150	120	71
ガラス転移点 (°C)		50	53	53	53
融点 (°C)		156	157	158	153
引張破断歪 (%)		10	6	11	200
引張破断強度 (kgf/cm <sup>2</sup> )		390	490	420	350
引張初期弾性率 (kgf/cm <sup>2</sup> )		13000	22000	19000	6200

[0161]

[Table 3]

項 目		実施例 5	実施例 6	実施例 7	実施例 8
L-LD (重量%)		68	68	68	68
D-LD (重量%)		2	2	2	2
ポリ エ ー テ ル エ ス テ ル	組成	SeA/EG/PPG	SeA/PG/PPG	SeA/EG/PEG	SeA/EG/PEG
	組成比(モル%)	100/30/70	100/75/35	100/30/70	100/30/70
	ポリエーテルのMw	3000	3000	2000	2000
	量 (重量%)	30	30	30	30
	Mw $\times 10^{-3}$	32	35	33	58
Mw $\times 10^{-3}$		88	120	88	130
ガラス転移点 (°C)		50	53	50	50
融点 (°C)		156	152	156	152
引張破断歪 (%)		10	15	120	94
引張破断強度 (kgf/cm <sup>2</sup> )		360	330	200	210
引張初期弾性率 (kgf/cm <sup>2</sup> )		18000	12000	7100	6800

[0162]

[Table 4]



項 目		実施例 9	実施例 10	実施例 11	実施例 12
L-LD (重量%)		68	68	68	68
D-LD (重量%)		2	2	2	2
ポリ エー テ ル ポ リ エ ス テ ル	組成	AA/EG/PPG	SA/EG/PPG	AA/EG/PPG /PMDA	AA/EG/PPG /PMDA
	組成比(モル%)	100/75/35	100/75/35	100/75/35	100/75/35
	ポリエーテルポールの Mw	3000	3000	3000	3000
	量 (重量%)	30	30	30	30
	$Mw \times 10^{-3}$	40	39	90	40
$Mw \times 10^{-3}$		110	120	130	110
ガラス転移点 (°C)		50	50	53	50
融点 (°C)		152	152	150	152
引張破断歪 (%)		27	29	43	31
引張破断強度 (kgf/cm <sup>2</sup> )		280	270	220	330
引張初期弾性率 (kgf/cm <sup>2</sup> )		10000	10000	9100	10000

[0163]

[Table 5]

項 目		実施例 1 3	実施例 1 4
L-LD (重量%)		6 8	6 8
D-LD (重量%)		2	2
ポリ エ ー ス テ ル	組成	SeA/EG/EbP	SeA/EG/EbP/tEbP
	組成比(モル%)	100/75/35	100/47/ 46/7
	ポリエーテルポリオール のMw	4 0 2 5	EbP : 4 0 2 5 tEbP : 3 0 0 0
	量 (重量%)	3 0	3 0
	$Mw \times 10^{-3}$	4 4	5 9
	$Mw \times 10^{-3}$	8 3	1 3 0
ガラス転移点 (°C)		5 0	5 1
融点 (°C)		1 5 2	1 5 2
引張破断歪 (%)		3 2	4 0
引張破断強度 (kgf/cm <sup>2</sup> )		3 1 0	2 9 0
引張初期弾性率 (kgf/cm <sup>2</sup> )		1 0 0 0 0	1 0 0 0 0

[0164]

[Table 6]

項 目		比較例 1	比較例 2
原 料	L-LD (wt%)	1 0 0	7 0
	D-LD (wt%)	0	0
	ポリエステル (wt%)	—	トーン 3 0
	$Mw \times 10^{-3}$	—	—
	$Mw \times 10^{-3}$	2 7 3	2 2 3
ガラス転移点 (°C)		5 7	4 7
融点 (°C)		1 5 8	1 4 9
引張破断歪 (%)		3	3 0
引張破断強度 (kgf/cm <sup>2</sup> )		5 0 0	2 5 0
引張初期弾性率 (kgf/cm <sup>2</sup> )		1 6 0 0 0	1 3 0 0 0

[0165]

[Effect of the Invention] This invention can offer manufacture \*\*\*\*\* of the lactic-acid system polyether polyester of biodegradability which has sufficient amount of macromolecules, thermal resistance, and thermal stability, and has the rigidity and flexibility according to an application, and transparency.

[Translation done.]

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TECHNICAL FIELD

[Industrial Application] This invention relates to the manufacture approach of the lactic-acid system copolymer of the amount of macromolecules in which various fabrication useful into shaping resin, a sheet film material, coating resin, ink resin, adhesives resin, the resin for laminations to paper, and a foaming resin ingredient is possible.

[0002] It is lactide system polyether polyester of the amount of giant molecules, and the lactic-acid system copolymer made by the manufacture approach of this invention has biodegradability, and the fabrication by various approaches, such as blow molding, extrusion molding, injection molding, inflation molding, laminate molding, press forming, and extrusion-foaming shaping, is possible for it, and it can fabricate it using the shaping equipment for general-purpose resin.

[0003] The lactic-acid system copolymer made by the manufacture approach of this invention specifically As above-mentioned various the application for sheets, for example, object, to a tray, a cup, a pan, a blister, etc., as a film As a blow molding article, it is [ material / a disposable diaper, sanitary items, an artificial kidney, a suture, / medical-application ] [0004] as sanitary goods to a shampoo bottle, a cosmetics bottle, a drink bottle, an oil container, etc. in bags, such as a wrap film, food packing, other general packages, a garbage bag, a register bag, a general specification bag, and a heavy bag. As agricultural materials, into the multifilm for agriculture, the gradual release agent sheet of agricultural chemicals, an anti-bird network, a care-of-health sheet, a plant pot, a fruits bag, etc., as fishery materials To a fishing net, a dried-seaweed culture network, a fishing line, a ship bottom paint, etc., as an injection-molded product It is [0005] to the miscellaneous-goods article for days, stationery, etc., such as a shank of the heart of golf tea, false food, and a cotton swab, the rod of a candy, a brush, a gear-tooth brush, a glass syringe, a pan, a cup, a comb, and a razor, a cassette of a tape, a disposable spoon fork, and a ball-point. In addition to this as a lamination to paper, it is used for a binding tape, a prepaid card, a balloon, a panty hose, a hair cap, sponge, a cellophane tape, an umbrella, a raincoat, a plastic glove, a hair cap, a rope, a nonwoven fabric, a tube, a foaming tray, foaming shock absorbing material, shock absorbing material, packaging, hot melt adhesive, the filter of a cigarette, a T-shirt, etc. useful at a tray, a cup, a pan, a megaphone, etc.

[Translation done.]

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## PRIOR ART

[Description of the Prior Art] In recent years, research which is going to utilize widely the lactic-acid system polymer which has the outstanding biodegradability from an environmental problem etc. as a general-purpose polymer is done briskly, and many researches on the manufacture approach and patent application are made. However, conventionally, if it is hard to say that the copolymer of the polylactic acid which is the polymer of a well-known lactic acid or lactide or a lactic acid, and other monomers has the sufficiently satisfactory engine performance in a moldability, transparency, and thermal resistance and a special application is removed, there is a trouble in using as general-purpose resin, and much of amelioration of these lactic-acids system polymer is expected.

[0007] The block copolymer which consists of a polylactic acid segment and a polypropylene-glycol segment, its manufacture approach, a copolymer film, and copolymerization are indicated by JP,1-108226,A. Moreover, the copolymer of a lactic acid and a polyethylene glycol is indicated by JP,1-108226,A.

[0008] However, by these approaches, if the amount of copolymerization of polyol is made [ many ], molecular weight will become extremely low. For example, in the example 4 of JP,1-108226,A, when 10 % of the weight of polypropylene glycols of number average molecular weight 4,000 is copolymerized with lactide, as for the number average molecular weight of the copolymer obtained, only the copolymer of 30,000 is obtained. Moreover, it is difficult to lose the reinforcement of the copolymer which consists of these lactic acids and polyol, if about 40 degrees C is exceeded, and to be equal to the use in an elevated temperature as common mold goods other than medical application.

[0009] As a copolymer of lactide and aliphatic series polyester, the polymerization of the epsilon-caprolactone is carried out beforehand, a homopolymer is obtained, and the approach of block-copolymerization-izing lactide further is indicated by JP,63-145661,A.

[0010] However, by the approach of block-copolymerization-izing lactide to a Pori epsilon-caprolactone, the obtained copolymer carries out nebula opacification. It is hard to dissolve the Pori epsilon-caprolactone block and polylactic acid block in a copolymer, and this is considered for becoming cloudy reflecting the high crystallinity which the aliphatic series system polyester of a Pori epsilon-caprolactone chain generally has. Moreover, in spite of the comparatively high glass transition point by differential thermal analysis, it is a flexible property at a room temperature.

[0011] When these Prior arts were summarized and sufficient reinforcement, thermal resistance, and thermal stability were given, it lacked in flexibility and transparency, and since it would become scarce at reinforcement, thermal resistance, and thermal stability if sufficient flexibility and transparency are given, the polymer equipped with the property it can be satisfied with still using it as ingredient resin, such as a film sheet, of a property enough was not obtained.

[0012] Plasticization of the polymer by the additive can be considered as these physical-properties improvement means. However, an improvement of the physical properties by these plasticization approach also has problems, such as reduction, disappearance, etc. of the plasticization effectiveness according [ the lactide which is a plasticizer ] to disappearance, out of a polymer at exsorption and a list during storage of the adhesion contamination problem to the equipment in the production process by a

heat-resistant fall and sublimation scattering of lactide, and a product, or use, when the lactide which is a residual monomer is used as a plasticizer as a means of plasticization.

[0013] Moreover, also when the plasticizer for the polymers of general well-known common use was added, a lot of plasticizer addition for plasticization was needed, the problem of the bleed out of a plasticizer is unescapable, problems, such as disappearance of the plasticization effectiveness of a under [ storage ], were not solved, and the polymer equipped with a satisfying enough property as a polymer for wrapping was not obtained. Moreover, it was not easy to obtain the lactic-acid system polymer of the amount of macromolecules, either.

[Translation done.]

**\* NOTICES \***

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**EFFECT OF THE INVENTION**

[Effect of the Invention] This invention can offer manufacture \*\*\*\*\* of the lactic-acid system polyether polyester of biodegradability which has sufficient amount of macromolecules, thermal resistance, and thermal stability, and has the rigidity and flexibility according to an application, and transparency.

[Translation done.]

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TECHNICAL PROBLEM

[Problem(s) to be Solved by the Invention] Therefore, the technical problem which this invention tends to solve is to offer the manufacture approach of the lactic-acid system polyether polyester of biodegradability of having sufficient amount of macromolecules, thermal resistance, and thermal stability, and having the rigidity and flexibility according to an application, and transparency.

[Translation done.]



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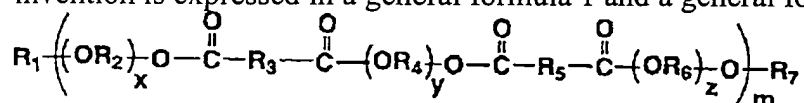
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## MEANS

[A means to solve a technical problem] Wholeheartedly this inventions as a result of examination that such a technical problem should be solved Lactide, By making the polyether polyester which consists of the polyether polyol component and polyol component which consist of various configuration rates, and a dicarboxylic acid component react as an indispensable component Moreover, by making the polyether polyester which consists of lactide, and the polyether polyol component and polyol component which consist of various configuration rates, a dicarboxylic acid component, the polyvalent carboxylic acid of three or more organic functions and/or its acid anhydride react as an indispensable component Controlling the decomposition to a monomer, having the molding temperature of the large range, and having the thermal stability at the time of high reinforcement and shaping at the time of shaping, [0016] Moreover, that various lactic-acid system copolymers of even the resin which has the flexible high property of the toughness which has high molecular weight easily, tears when it is processed in the shape of a film from hard resin with a high glass transition point and the high melting point, is excellent in reinforcement, and cannot carry out a chip box crack easily changing the rate of this polyether polyester into arbitration in case copolymerization-ization is performed with lactide, or by changing the class of polyether polyester can be manufactured, [0017] That is, it finds out that the lactic-acid system copolymer of the amount of macromolecules of resolvability which has sufficient reinforcement which can be used as general-purpose resin, and the thermal stability at the time of shaping, and has the rigidity according to an application, transparency, and flexibility can be manufactured, and came to complete this invention.

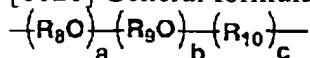
[0018]

[Elements of the Invention] namely, the polyether polyester (A) which has the repeat unit as which this invention is expressed in a general formula 1 and a general formula 1 -- [Formula 3]



[0019] (mx, y, and z are one or more integers among a formula.) As for R1 and R7, the sum totals of a methylene carbon number of hydrogen or a hydrocarbon group, and R3 and R5 are two or more aliphatic hydrocarbon radicals, and R2, R4, and R6 have the chemical structure of an alkylene group or a general formula 2.

[0020] General formula 2 [Formula 4]



(Among a formula, R8, R9, and R10 are alkylene groups, and c is a, b, 0, or one or more integers, and a+b+c is one or more integers.)

It is the manufacture approach of the lactic-acid system polyether polyester characterized by carrying out copolymerization of the lactide (B) under existence of a polymerization catalyst (C).

[0021] Moreover, the manufacture approach of the lactic-acid system polyether polyester of this

invention being polyether polyester with which macromolecule quantification of this polyether polyester (A) was carried out by the polyvalent carboxylic acid (and/or, the acid anhydride) (D) of three or more organic functions, and this polyether polyester (A) are characterized by being 999 / 1 - 200/800 by the mole ratio of the ether group in polyether polyester, and an ester group.

[0022] Moreover, the manufacture approach of the lactic-acid system polyether polyester of this invention is characterized by the weight ratios of polyether polyester (A) and lactide (B) being 60 / 40 - 5/95.

[0023] furthermore, this invention -- polyether polyol (E), polyol (F), and dicarboxylic acid (G) -- and/or, it is the manufacture approach of the polyether polyester (H) obtained by making the polyvalent carboxylic acid of three or more organic functions, and/or its acid anhydride (D) react, and the lactic-acid system polyether polyester characterized by carrying out copolymerization of the lactide (B) under existence of a polymerization catalyst (C).

[0024] The manufacture approach of the lactic-acid system polyether polyester of this invention is characterized by the weight ratios of that polyether polyester (H) is [ the mole ratios of the ether group in polyether polyester (H) and an ester group ] 999 / 1 - 200/800, this polyether polyester (H), and lactide (B) being 60 / 40 - 5/95.

[0025] Furthermore, polyether polyol (E) is characterized by being one or more kinds of polyether polyols chosen from a polyethylene glycol, a polypropylene glycol, and the Pori (oxyethylene-oxypropylene) glycol that is the copolymer of ethyleneoxide and propylene oxide especially in detail. Moreover, polyol (F) is characterized by being one or more kinds of diols chosen from ethylene glycol and propylene glycol.

[0026] Moreover, a carboxylic acid (G) is characterized by being one or more kinds of dicarboxylic acid chosen from a succinic acid, an adipic acid, a sebacic acid, its acid anhydride and those methyl ester, and ethyl ester especially in detail.

[0027] furthermore, this invention -- polyol (F) and dicarboxylic acid (G) -- and/or, the manufacture approach of the polyester which consists of the polyvalent carboxylic acid of three or more organic functions and/or its acid anhydride (D), the polyether polyester (I) obtained by carrying out the ester exchange reaction of the polyether polyol (E), and the lactic-acid system polyether polyester characterized by carrying out copolymerization of the lactide (B) under existence of a polymerization catalyst (C) is also included.

[0028] As for the manufacture approach of the lactic-acid system polyether polyester of this invention, polyether polyester (I) is further characterized by to be 999 / 1 - 200/800 and the weight ratios of polyether polyester (I) and lactide (B) being 60 / 40 - 5/95 by the mole ratio of the ether group in polyether polyester, and an ester group.

[0029] Moreover, it is that polyether polyol (E) is one or more kinds of polyether polyols chosen from a polyethylene glycol, a polypropylene glycol, and the Pori (oxyethylene-oxypropylene) glycol that is the copolymer of ethyleneoxide and propylene oxide, and [0030] in detail. Polyol's (F's)'s being one or more kinds of diols chosen from ethylene glycol and propylene glycol and dicarboxylic acid (G) are the manufacture approaches characterized by being one or more kinds of dicarboxylic acid chosen from a succinic acid, an adipic acid, a sebacic acid, its acid anhydride and those methyl ester, and ethyl ester.

[0031] Furthermore, the polyester which this invention makes dicarboxylic acid (G) react with polyol (F), and is obtained, The polyether polyester obtained by making the polyvalent carboxylic acid (and/or, the acid anhydride) of three or more organic functions react to the polyether polyester which polyether polyol (E) is made to react and is obtained (J), The manufacture approach of the lactic-acid system polyether polyester characterized by carrying out copolymerization of the lactide (B) under existence of a polymerization catalyst (C) is also included.

[0032] The manufacture approach of the lactic-acid system polyether polyester of this invention is characterized by the weight ratios of that this polyether polyester (J) is [ the mole ratios of the ether group in polyether polyester and an ester group ] 999 / 1 - 200/800, polyether polyester (J), and lactide (B) being 60 / 40 - 5/95.

[0033] Furthermore, it is that polyether polyol (E) is one or more kinds of polyether polyols chosen from

a polyethylene glycol, a polypropylene glycol, and the Pori (oxyethylene-oxypropylene) glycol that is the copolymer of ethyleneoxide and propylene oxide, and [0034] in detail. Polyol's (F's)'s being one or more kinds of diols chosen from ethylene glycol and propylene glycol and dicarboxylic acid (G) are characterized by being one or more kinds of dicarboxylic acid chosen from a succinic acid, an adipic acid, a sebacic acid, its acid anhydride and those methyl ester, and ethyl ester.

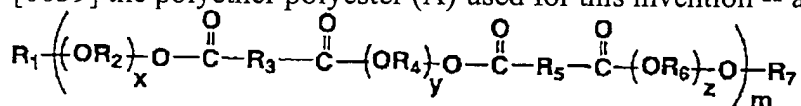
[0035] Below, this invention is explained at a detail. Sequential explanation is given about the lactide and polyether polyester which are used by this invention, dicarboxylic acid, polyether polyol, polyol, the polyvalent carboxylic acid of three or more organic functions, its acid anhydride, etc.

[0036] The lactide used by this invention is the compound which cyclic-ester-ized the lactic acid between dyad, and is a monomer which has a stereoisomer. That is, L-lactide which consists of two L-lactic acid, D-lactide which consists of a D-lactic acid, and the MESO-lactide which consists of L-lactic acid and a D-lactic acid exist in lactide.

[0037] Although the copolymer only containing L-lactide or D-lactide is crystallized and high-melting is obtained, by the manufacture approach of the lactic-acid system copolymer of the amount of macromolecules of this invention, the desirable resin property according to an application is realizable by combining these three sorts of lactide.

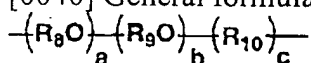
[0038] In order for the lactide used in order to discover a high thermal property in this invention to discover a thermal property with what [ desirable still higher ] contains 75% or more for L-lactide among the total lactide, as for lactide, what contains 90% or more for L-lactide among the total lactide is desirable.

[0039] the polyether polyester (A) used for this invention -- a general formula 1 -- [Formula 5]



(mx, y, and z are one or more integers among a formula.) As for R1 and R7, the sum totals of a methylene carbon number of hydrogen or a hydrocarbon group, and R3 and R5 are two or more aliphatic hydrocarbon radicals, and R2, R4, and R6 have the chemical structure of an alkylene group or a general formula 2.

[0040] General formula 2 [Formula 6]



(Among a formula, R8, R9, and R10 are alkylene groups, and c is a, b, 0, or one or more integers, and a+b+c is one or more integers.)

[0041] Furthermore, in detail, as for R1 and R7 in a general formula 1 and a general formula 2, the sum total of a methylene carbon number of hydrogen or the hydrocarbon group of carbon numbers 1-4, and R3 and R5 is the aliphatic hydrocarbon radical of 2-12, and, as for carbon numbers R2, R4, R6, R8, R9, and R10, the carbon number of an alkylene principal chain has the alkylene group of 2-15, or the chemical structure of a general formula 2.

[0042] Specifically, the hydrocarbon group more than divalent [ of methylene ethylene, trimethylene [- (CH2) 3-], propylene, tetramethylen, and [CH3CH2C(CH2-)-3] [-CH(CH2-)-2 grade ] especially ethylene, a propylene, and [CH3CH2C(CH<SUB>2</SUB>-)-3] [-CH (CH2-)-2 are mentioned.

[0043] the polyether polyester from which polyether polyester (H) is obtained by the dehydration and the deglycol reaction of polyether polyol, polyol, and dicarboxylic acid -- or it is polyether polyester obtained by the dehydration and the deglycol reaction of polyether polyol, polyol, dicarboxylic acid, and the polyvalent carboxylic acid (and/or, the acid anhydride) of three or more organic functions.

[0044] the polyether polyester with which the ester interchange of the polyether polyester (I) used for this invention was carried out, and polyol and dicarboxylic acid were obtained in dehydration, and the polyester and polyether polyol which is made to carry out a deglycol reaction and is obtained -- or it is polyether polyester which was made to carry out the ester interchange of polyol, dicarboxylic acid, and

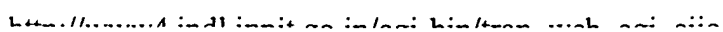
[0045] The polyether polyester (J) used for this invention is polyether polyester which carried out the ester interchange of the polyvalent carboxylic acid (and/or, the acid anhydride) of three or more organic functions to the polyether polyester which was made to carry out the ester interchange of polyol and the dicarboxylic acid, and was obtained [ dicarboxylic acid ] in dehydration and the polyester which is made to carry out a deglycol reaction and is obtained, and polyether polyol, it was made to react to it, and was obtained.

[0047] Furthermore, it is more desirable that the polyether polyester (A) from a viewpoint of the solubility of polyether polyester and the ease of manufacture, (H), (I), or (J) is 10,000 to about 100,000 in 20,000-200,000, and number average molecular weight at weight average molecular weight.

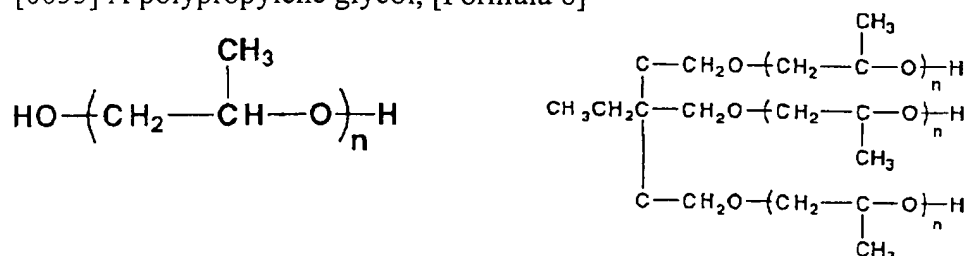
[0049] Moreover, the thermal resistance of the lactic-acid system copolymer of lactide (E) and polyether polyester ((A), (H), (I), (J)) comparatively obtained as it is 40 / 60 - 95/5 becomes high. furthermore, (A)/((A), (H), (I) or, and (J)) -- comparatively -- 50/50- the thermal resistance of the amount lactide system copolymer of macromolecules obtained as it is 80/20 becomes higher and is desirable.

[0051] Although especially the aliphatic series dicarboxylic acid component in the polyether polyester used by this invention is not specified, it is desirable that it is the aliphatic series dicarboxylic acid (and those acid anhydrides), those methyl ester, and ethyl ester of the carbon atomic numbers 4-14. A succinic acid, an adipic acid, a sebacic acid, cyclohexane dicarboxylic acid and those methyl ester, ethyl ester, a succinic anhydride, a hexa hydro phthalic acid, etc. are desirable especially.

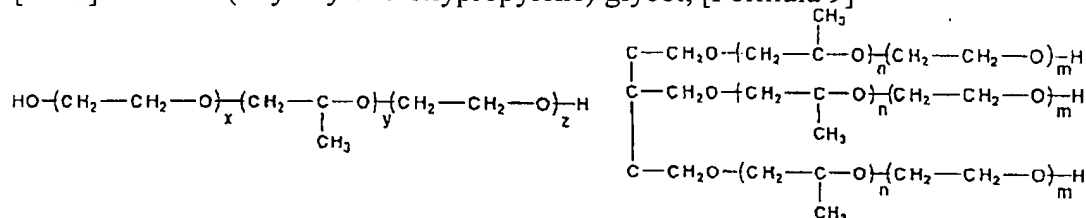
[0053] It is [0054], when a polyethylene glycol, a polypropylene glycol, the Pori (oxyethylene-oxypropylene) glycol that is the copolymer of ethyleneoxide and propylene oxide, Pori 1, 2-butylene glycol, Pori 1, 4-butylene glycol, and poly neopentyl glycol are desirable and consider biodegradability, versatility, etc. further especially, although a class will not be asked diol, triol, and more than it especially about the polyether polyol component in the polyether polyester used for this invention if it is polyether polyol. A polyethylene glycol, [Formula 7]



[0055] A polypropylene glycol, [Formula 8]



[0056] The Pori (oxyethylene-oxypropylene) glycol, [Formula 9]



Especially \*\* is desirable.

[0057] Moreover, as for the molecular weight of polyether polyol (A), it is desirable that it is 200-200,000 in weight average molecular weight. Furthermore, it is more desirable that the viewpoint of biodegradability to polyether polyol (A) is 200 to about 10,000 in weight average molecular weight.

[0058] Although a class will not be asked about the polyol component in polyether polyester especially if it is polyol, the polyol of 2-15 has a desirable carbon number especially, and it is specifically ethylene glycol, propylene glycol, a butylene glycol, pentane polyol, hexamethylene glycol, octane polyol, neopentyl glycol, cyclohexane dimethanol, a xylene glycol, and [0059]. a diethylene glycol, triethylene glycol, dipropylene glycol, and a jib -- hydrogenation bisphenol A, such as tongue diol, 3-hydroxy pivalyl pivalate, a glycerol, and pentaerythritol, is mentioned.

[0060] Although a class is not asked especially concerning the polyvalent-carboxylic-acid component of three or more organic functions, trimesic acid, propane tricarboxylic acid, trimellitic acid, trimellitic anhydride, pyromellitic acid, pyromellitic dianhydride, benzophenone tetracarboxylic acid, 1 and 3, 5-cyclohexane tricarboxylic acid, cyclohexane tetracarboxylic acid, a cyclohexane tetracarboxylic acid anhydride, etc. are mentioned especially.

[0061] Especially trimellitic anhydride and pyromellitic dianhydride are desirable. The above-mentioned polyvalent carboxylic acid can be mixed and used if needed. Especially, the amount of the polyvalent carboxylic acid used is explained below.

[0062] Polyether polyol (E), polyol (F), and dicarboxylic acid (G), In obtaining polyether polyester (H) dehydration and by carrying out a deglycol reaction, the polyvalent carboxylic acid (and/or, the acid anhydride) (D) of three or more organic functions The molecular weight of the lactic-acid system copolymer of the amount of macromolecules of the polyvalent carboxylic acid (D) of three or more organic functions comparatively obtained to a dicarboxylic acid (G) component as 0.001 - five-mol % is desirable and it is 0.01 - one-mol %, and flexibility become high.

[0063] And a deglycol reaction is carried out and it is obtained. polyol (F), dicarboxylic acid (G) and the polyvalent carboxylic acid of three or more organic functions, and/or its acid anhydride (D) -- dehydration -- with polyester In obtaining polyether polyester (I) by carrying out the ester exchange reaction of the polyether polyol (E) The molecular weight of the lactic-acid system copolymer of the amount of macromolecules of the polyvalent carboxylic acid (D) of three or more organic functions comparatively obtained to a dicarboxylic acid (G) component as 0.001 - five-mol % is desirable and it is especially 0.01 - one-mol %, and flexibility become high.

[0064] It is [0065] when obtaining polyether polyester (J) by carrying out the ester exchange reaction of the polyvalent carboxylic acid of three or more organic functions, and/or its acid anhydride (D) to polyol

(F) and the polyether polyester which was made to carry out the ester interchange of the dicarboxylic acid (G), and was obtained [ dicarboxylic acid ] in dehydration and the polyester which is made to carry out a deglycol reaction and is obtained, and polyether polyol (E). 0.01 - 5 % of the weight is desirable to a component in the polyether polyester of the polyvalent carboxylic acid (D) of three or more organic functions which was made to carry out the ester interchange of the dicarboxylic acid (G) with polyol (F) comparatively, and was obtained in dehydration and the polyester which is made to carry out a deglycol reaction and is obtained, and polyether polyol (E), and the molecular weight of the lactic-acid system copolymer of the amount of macromolecules obtained as it is 0.1 - 1 % of the weight, and flexibility become high especially at it.

[0066] Branched chain is introduced by using the polyvalent carboxylic acid (D) of these three or more organic functions. The closure of the end of the effectiveness that molecular weight distribution are expanded to the increase list of the molecular weight of a copolymer, and the generated copolymer is carried out, and thermal stability increases, or a metal reacts to the carboxyl group of one or more functional groups of polyvalent carboxylic acid. When a polymer ionomer-izes, macromolecule quantification is easily possible and the lactic-acid system copolymer which can be fabricated on the film and sheet which have the outstanding physical properties can be manufactured.

[0067] If the lactic-acid system copolymer of the amount of macromolecules obtained by this invention is sheet-ized, from the sheet of high reinforcement to a flexible sheet can be obtained. Specifically, it is 500-50,000kg/cm<sup>2</sup> as hauling viscoelasticity. A sheet is obtained.

[0068] Macromolecule quantification can be further carried out by making it react with one or more sorts of compounds chosen from the group which changes to the obtained lactic-acid system copolymer from hydroxy acid, carboxylic anhydrides, isocyanates, and lactone. If these examples are given, for example as hydroxy acid, a citric acid, a tartaric acid, a malic acid, etc. will be an acetic anhydride, a succinic anhydride, a propionic anhydride, phthalic anhydride, a maleic anhydride, a butyric anhydride, an isobutyric anhydride, trimellitic anhydride, pyromellitic dianhydride, etc. as carboxylic anhydrides.

[0069] moreover -- isocyanate -- a kind -- \*\*\*\*\* -- phenyl -- isocyanate -- hexamethylene diisocyanate -- toluene 2,4-diisocyanate -- naphthalene - one -- five - diisocyanate -- a triphenylmethane color - four -- four -- ' -- four -- " - tri-isocyanate -- etc. -- lactone -- a kind -- \*\*\*\*\* -- beta - a caprolactone -- epsilon - a caprolactone -- beta - a butyrolactone -- delta - a valerolactone -- etc. -- mentioning -- having .

[0070] In order to make these compounds react to a lactic-acid system copolymer further, it mixes in the reactant after the polymerization reaction of a lactic-acid system copolymer is completed, and changes into the lactic-acid system copolymer which carried out stirring mixing in the state of short-time melting, or was obtained by the polymerization, and these may be added and melting mixing may be carried out. Moreover, after making it dissolve in a common solvent and making both lactic-acid system copolymer and these compounds heat and react to it, you may obtain by reprecipitation or degassing.

[0071] Especially, about hydroxy acid, carboxylic anhydrides, and lactone, the approach of adding these to the obtained lactic-acid system copolymer, and making it react to it under reduced pressure after melting mixing is desirable, and, thereby, reaction time becomes quick.

[0072] Generally 60 degrees C - 240 degrees C of temperature made to mix and react are 80 degrees C - 190 degrees C preferably. Moreover, on the occasion of a reaction, ester polymerization catalysts, such as N.N-dimethylaniline, tin octanoate, a dibutyl tin JIRAU rate, and isopropyl titanate, or an urethane catalyst can be used. Whenever [ reduced pressure / in the case of a reaction ] is 3 or less Torrs still more preferably 10 or less Torrs preferably 100 or less Torrs.

[0073] 0.001 % of the weight - 5% of the weight of a lactic-acid system copolymer is desirable still more desirable, and the amount of hydroxy acid, carboxylic anhydrides, isocyanates, and the lactone used is 0.001 % of the weight - 0.1 % of the weight. The used hydroxy acid, carboxylic anhydrides, isocyanates, and the lactone of this approach are desirable also at the point that it combines with the end group of a lactic-acid system copolymer, and the decomposition to the monomer of the polymer by heat can be prevented.

[0074] It is desirable to use a polymerization catalyst (C) for manufacture of this invention. As a

polymerization catalyst (C) used by this invention, metals, such as the tin generally known also as the polymerization catalyst of cyclic ester and an ester interchange catalyst, zinc, lead, titanium, a bismuth, a zirconium, and germanium, and the derivative of those are mentioned. About especially these derivatives, an organic metal compound, a carbonate, an oxide, and a halogenide are desirable.

Specifically, tin octanoate, chlorination tin, a zinc chloride, zinc acetate, a lead oxide, lead carbonate, a titanium chloride, alkoxy titanium, the germanium dioxide, and the zirconium dioxide are suitable.

[0075] The amount of the polymerization catalyst (C) used has 0.005 - 0.2 desirable % of the weight to the sum total of lactide (B), polyether polyester ((A), (H), (I), or (J)), and/or the polyvalent carboxylic acid (D) of three or more organic functions, its rate of polymerization is fully quick, and especially in order to lessen coloring of the obtained lactide system polyether polyester, its 0.01 - 0.1 % of the weight is desirable.

[0076] Dehydration and in case it carries out a deglycol reaction and polyether polyester is manufactured, a catalyst can also be used for the polyvalent carboxylic acid (D), dicarboxylic acid, and polyol of three or more organic functions. As a catalyst used by this invention, as for each catalyst generally known as an esterification catalyst, metallic compounds organic [ , such as tin, zinc, lead titanium, antimony, a cerium, germanium, cobalt manganese, iron, aluminum, magnesium, calcium, and strontium, / of at least a kind of metal ] or inorganic are mentioned usable.

[0077] For example, metaled alkoxide, an organic-acid salt, a chelate, oxide, etc. are used, and compounds, such as the organic compound of titanium, for example, titanic-acid alkyl ester, titanium oxy-acetylacetonate, and a titanium oxalate, are especially useful. As amount of the catalyst used, 0.001 - 0.5 % of the weight is desirable to the polyvalent carboxylic acid (D), the dicarboxylic acid, and the polyol sum total of three or more organic functions. A rate of polymerization is fully quick, and especially in order to lessen coloring of the obtained lactide system polyether polyester, 0.01 - 0.1 % of the weight is desirable.

[0078] Next, the concrete manufacture approach of this invention is explained. In case polyether polyester (A) and the polyvalent carboxylic acid (and/or, the acid anhydride) (D) of three or more organic functions are made to react, when it carries out under reduced pressure, a reaction goes on promptly and is desirable after melting mixing. the reaction which mixes lactide (E) with the macromolecule quantification polyether polyester obtained by the reaction, and performs a polymerization -- mixture -- warming -- melting is carried out or a polymerization catalyst (F) is added for a reactant after dilution mixing with a solvent.

[0079] polyether polyol (E), polyol (F), and dicarboxylic acid (G) -- and/or, the polymerization with the polyvalent carboxylic acid (and/or, the acid anhydride) (D) of three or more organic functions performs the dehydration and the deglycol reaction which are the synthetic approach of common polyester. the reaction which mixes lactide (B) with the obtained polyether polyester (H), and performs a polymerization -- mixture -- warming -- melting is carried out or a polymerization catalyst (F) is added for a reactant after dilution mixing with a solvent.

[0080] polyol (F) and dicarboxylic acid (G) -- and/or, the polymerization with the polyvalent carboxylic acid (and/or, the acid anhydride) (D) of three or more organic functions performs the dehydration and the deglycol reaction which are the synthetic approach of common polyester. Next, the ester exchange reaction of the polyether polyol (E) is carried out to the obtained polyester under reduced pressure after melting mixing. the reaction which mixes lactide (B) with the obtained polyether polyester (I), and performs a polymerization -- mixture -- warming -- melting is carried out or a polymerization catalyst (F) is added for a reactant after dilution mixing with a solvent.

[0081] Polyol (F) and the polymerization with dicarboxylic acid (G) perform the dehydration and the deglycol reaction which are the synthetic approach of common polyester. Next, the ester exchange reaction of the polyether polyol (E) is carried out to the obtained polyester under reduced pressure after melting mixing. Next, the ester exchange reaction of the obtained polyether polyester and the polyvalent carboxylic acid (and/or, the acid anhydride) of three or more organic functions is carried out under reduced pressure after melting mixing.

[0082] the reaction which mixes lactide (B) with the obtained polyether polyester (J), and performs a

polymerization -- mixture -- warming -- melting is carried out or a polymerization catalyst (F) is added for a reactant after dilution mixing with a solvent. When the dehydration and the deglycol reaction which are the synthetic approach of common polyester here are described briefly, as for dehydration, it is desirable to carry out under an inert gas ambient atmosphere for 150-250 degrees C and 3 to 16 hours.

[0083] The deglycol reaction is gradually carried out to reduced pressure, and, finally it is desirable under reduced pressure of 5 or less Torrs to carry out at 170-260 degrees C for 2 to 16 hours. thus, the polyether polyester ((A), (H), (I) or, and (J)) and lactide (E) which were obtained -- warming -- when carrying out melting or adding a polymerization catalyst (F) after mixing by solvent addition, a polymerization system is made to homogeneity as it is more than the melting point of lactide, a quick rate of polymerization is obtained, and polymerization temperature is desirable.

[0084] The polymerization temperature in a non-solvent system is more than the melting point of lactide, is desirable on the balance of a polymerization, and can prevent coloring of the lactide system polyether polyester accompanying a decomposition reaction. [ of the temperature of 185 degrees C or less ] The melting point of lactide is near 100 degree C, and can prevent the temperature of 185 degrees C or less of 100 degrees C or more, the fall of the molecular weight of the lactide system polyether polyester accompanying [ 145-180 degrees C is desirable on the balance of a polymerization still more preferably, and ] a decomposition reaction, and coloring.

[0085] moreover, the thing for which all reactions are performed under the dry inert gas ambient atmosphere in order to prevent decomposition of lactide, and coloring -- it is desirable. It carries out in the state of bubbling under nitrogen and an argon gas ambient atmosphere especially. It is desirable that the polyether polyol and polyether polyester it is incomparable in a raw material also remove moisture to coincidence, and dry it by reduced pressure drying etc.

[0086] Moreover, since lactide can be dissolved in a solvent, a polymerization can be carried out using a solvent and benzene, toluene, ethylbenzene, a xylene, a cyclohexanone, a methyl ethyl ketone, isopropyl ether, etc. are mentioned as an example of a solvent, for example.

[0087] The thing which used the usual polymerization iron pot and to do for batch manufacture is also possible for the lactic-acid system copolymer of the amount of giant molecules of this invention. Furthermore, improvement in product quality, yield, and productivity can be aimed at by the continuation manufacture approach. As the continuation manufacture approach, the stirring type reactor connected with two or more serials, a static mixer, and/or a horizontal-type reactor are used for all or a part of the reaction processes.

[0088] Use of the stirring type reaction vessel which changed the stirring approach with advance of a reaction and was connected with the serial of two or more tubs in which efficient stirring and temperature control are possible as a stirring type reactor connected with two or more serials is desirable.

[0089] The static mixer which says here is static mixing equipment which the amount of moving part is not to the mixed equipment which has an agitator, namely, does not have an agitator, flow divides, and a flow direction converts or reverses by the mixing element the amount of [ which was fixed in tubing ] moving part is not specifically, and the mixed equipment which mixes a solution says by repeating division, conversion, and reversal for flow in a lengthwise direction and a longitudinal direction. Moreover, of course, an extruder [ like one shaft used for reactive processing or a biaxial extruder ] whose horizontal-type reactor is says the thing of the biaxial reactor through which it can circulate.

[0090] Even if a solvent etc. is used for these continuation manufacture approach, it can carry out a polymerization. Although the lactic-acid system copolymer of the obtained amount of macromolecules has high melt viscosity the top where the melting point is high and it is hard to carry out a polymerization by this, it falls, stirring becomes easy and the viscosity of a polymerization system becomes easy to perform a polymerization by adding a solvent.

[0091] the polymerization equipment which the knockout pressure of a polymerization solution fell and had the interior equipment for heat carriers, and a baffle aiming at stirring for the purpose of the temperature control when continuation polymerization equipment equipped with especially the static



mixer was used -- equipment -- casual dress -- it does-izing and is effective. Since stirring is easy, a temperature control is easy, temperature is homogeneous in polymerization equipment, and the lactic-acid system copolymer of the amount of macromolecules with less coloring etc. is obtained.

[0092] Moreover, it is desirable to devolatilize under reduced pressure in order to remove the matter with the lactide, solvent, and odor which remained at the polymerization anaphase. The amount of residual lactide can be decreased according to this devolatilization process, and the preservation stability of the lactic-acid system copolymer of the obtained amount of macromolecules can be increased remarkably.

[0093] Residual lactide becomes hydrolysis by adhesion of moisture etc., or the cause of the welding by heat and is not desirable when the amount lactide system polyether polyester of macromolecules is used as a sheet film. Moreover, it disperses [ by sublimation ] from the film sheet produced commercially and is not desirable. As for the amount of residual lactide in the lactic-acid system copolymer of this invention, for this reason, it is desirable to carry out to 2 or less % of the weight. It carries out to 1 or less % of the weight still more preferably.

[0094] As the approach of concrete devolatilization, one shaft or a biaxial extruder, a thin film distillation machine, a pot mold decompression device, etc. perform. As conditions for devolatilization, the method of performing ejection is desirable under reduced pressure after a polymerization, heating. In order not to reduce the molecular weight of a lactic-acid system copolymer, as for devolatilization conditions, it is desirable that whenever [ 100-230 degrees-C and reduced pressure ] performs devolatilization time amount, and temperature performs it by 0.1 - 200Torr for 10 seconds to 10 minutes.

[0095] There is a method of performing ejection pelletizing or grinding a lactic-acid system copolymer, and heating it under reduced pressure after polymerization termination, as the other devolatilization approaches. 0.1 - 50Torr of temperature is [ devolatilization time amount / whenever / 60-200-degree-C and reduced pressure ] desirable in order not to reduce the molecular weight of a lactic-acid system copolymer in this case, either for 2 to 400 minutes.

[0096] In case the copolymer of this invention is manufactured, cyclic ester other than lactide (E) can be added further, and the lactic-acid system copolymer of the amount of macromolecules can also be made. Lactone can be added one to 20% of the weight especially for the purpose of elasticity-izing. About the cyclic ester added in addition to lactide, although there is especially no constraint, the annular dimerization object of hydroxy acids, such as glycolide, intramolecular lactide especially beta-butyrolactone, epsilon-caprolactone, gamma-valerolactone, g undecalactone, etc. are specifically mentioned. If the amount of lactone increases, a glass transition point and the melting point will become low, and flexibility will become high.

[0097] In case lactic-acid system copolymerization of this invention is formed on a sheet film etc., mixing addition of the organic system bulking agents, such as inorganic system bulking agents, such as a general filler, for example, talc, a calcium carbonate, a silica, clay, the diatom earth, and a pearlite, or wood flour, may be carried out. Moreover, stabilizers, such as ultraviolet ray absorbents, such as 2,6-di-tert-butyl-4-methylphenol (BHT), an anti-oxidant like a butylhydroxyanisol (BHA), salicylic acid derivatives, a benzophenone system, and a benzotriazol system, and phosphoric ester, and a carbodiimide, can be used, and the thermal stability at the time of shaping can be raised.

[0098] Although the lactic-acid system copolymer of this invention has sufficient plasticity independently and has a good moldability, when planning still higher fabrication nature and flexibility, it may add plasticizers, such as dioctyl adipate, a dioctyl sebacate, trioctyl trimellitate, a diethyl phthalate, a dioctyl phthalate, a polypropylene-glycol adipic acid, and adipic-acid butanediol.

[0099] Moreover, even if a coloring agent like surface active agents, such as ion systems, such as non-ion systems, such as lubricant, such as metallic soap, such as zinc stearate, magnesium stearate, and stearic acid calcium, mineral oil, a liquid paraffin, and ethylene screw stearyl AMAIDO, a glycerine fatty acid ester, and cane-sugar fatty acid ester, and an alkyl sulfonate, titanium oxide, and carbon black etc. adds, it does not interfere.

[0100] moreover, addition of organic system foaming agents, such as inorganic system foaming agents,

such as sodium bicarbonate and ammonium bicarbonate, an AZOJI carvone amide, azobisisobutyronitril, and sulfonylhydrazide, etc. -- or -- or it infiltrates foaming agents, such as a pentane, butane, and Freon, into the lactic-acid system copolymer of this invention in advance -- an extrusion process -- on the way -- it can also consider as foam by coming out and supplying directly in an extruder. Moreover, multilayering with paper, aluminum foil, or other resolvability polymer films is also possible by extrusion lamination, dry lamination, or co-extrusion.

[0101] The lactic-acid system polyether polyester copolymer obtained by this invention has good biodegradability, and even if it is discarded after being used for general-purpose resin, wrapping, etc., or discarded from a production process, it is useful to loss in quantity of trash. When abandoned all over soil and seawater, decomposition by hydrolysis, a microorganism, etc. is received and the reinforcement as resin deteriorates in several months, and by the time it does not maintain an appearance, it can decompose. Moreover, if compost is used, biodegradation will be carried out by the time it does not stop the original form further for a short period of time.

[Translation done.]

## \* NOTICES \*

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1. This document has been translated by computer. So the translation may not reflect the original precisely.
2. \*\*\*\* shows the word which can not be translated.
3. In the drawings, any words are not translated.

## EXAMPLE

[Example] This invention is not limited by these examples although an example and the example of a comparison explain this invention still more concretely below. In addition, especially, the polyether polyol in an example used the diol mold altogether, as long as it was unstated.

[0103] In addition, the determination of molecular weight was measured by the comparison with a polystyrene correlation sample with the GPC measuring device (Following GPC, abbreviation, the column temperature of 40 degrees C, tetrahydrofuran solvent). The differential-scanning-calorie meter (Following DSC and abbreviation) performed a glass transition point and the melting point. The temperature up of the Measuring condition was carried out from 20 degrees C to 220 degrees C by 10 degrees C / min.

[0104] Moreover, the Measuring condition of a tension test was performed by 50mm of initial sample length, and crosshead rate 40 mm/min. The test piece cut and measured the film of 250-micrometer thickness created under 160-180 degrees C, 200kg/cm<sup>2</sup>, and the conditions for 2 minutes with the hotpress machine in width-of-face [ of 15mm ] x die length of 80mm.

[0105] In addition, the above-mentioned device used the device described below.

GPC:TOSOH HLC-8020 (TOSOH CORP. make)

DSC:DSC 200 (the SEIKO electronic industry incorporated company make)

Tension test: Tensilon (Oriental energy machine incorporated company make)

[0106] [Example 1] Heating stirring was carried out, teaching polypropylene-glycol 45g of number average molecular weight 400, ethylene glycol 4.1g, and 25g of sebacic acids to 300ml 4 opening flask which attached an agitator, a rectifier, and gas installation tubing, and carrying out a temperature up to it in 150 degrees C to 10 degrees C /under nitrogen-gas-atmosphere mind in an hour.

[0107] The temperature up was carried out to 220 degrees C, distilling off the water to generate, after checking that the distillate of water has stopped, titanium isopropyl titanate 0.01g was added and the deglycol reaction was performed at 220 degrees C under reduced pressure of 0.5Torr(s) for 3 hours. it is [ polyester / which was obtained / polyether ] \*\*\*\*\* in number average molecular weight 18,000 and weight average molecular weight 33,000 -- it was liquefied.

[0108] It taught 200ml separable flask, and under nitrogen-gas-atmosphere mind, it fused, obtained polyether polyester 30g, L-lactide 68g, D-lactide 2g, and 175-degree C toluene 10ml were mixed for 0.25 hours, and 0.03g of tin octanoate was added. The generated copolymer was taken out after the 3-hour reaction.

[0109] The obtained lactic-acid system polyether polyester was transparent, and was number average molecular weight 55,000 and weight average molecular weight 93,000 from GPC. The glass transition point was observed by about 50 degrees C, and the melting point was about 156 degrees C. Moreover, \*\*\*\* fracture distortion was [ 390 kgf/cm<sup>2</sup> and the initial elastic modulus of \*\*\*\* of \*\*\*\* breaking strength ] 13,000 kgf/cm<sup>2</sup> 10%.

[0110] [Example 2] Heating stirring was carried out, teaching ethylene glycol 21g and 50g of sebacic acids to 300ml 4 opening flask which attached an agitator, a rectifier, and gas installation tubing, and carrying out a temperature up to it in 150 degrees C to 10 degrees C /under nitrogen-gas-atmosphere

mind in an hour. The temperature up was carried out to 220 degrees C, distilling off the water to generate, after checking that the distillate of water has stopped, titanium isopropyl titanate 0.01g was added and the deglycol reaction was performed at 220 degrees C under reduced pressure of 0.5Torr(s) for 3 hours. The obtained polyester (A-1) was the white solid-state of number average molecular weight 20,000 and weight average molecular weight 35,000.

[0111] Obtained polyester (A-1) 13g and polypropylene-glycol 60g of number average molecular weight 3,000 were taught to 200ml 4 opening flask which attached an agitator, a rectifier, and gas installation tubing, and the deglycol reaction was performed at 220 degrees C under reduced pressure of 0.5Torr for 3 hours. it is [ polyester / (A-2) / which was obtained / polyether ] \*\*\*\*\* in number average molecular weight 21,000 and weight average molecular weight 36,000 -- it was liquefied.

[0112] It taught 200ml separable flask, and under nitrogen-gas-atmosphere mind, it fused, obtained polyether polyester 10g, L-lactide 85g, D-lactide 5g, and 175-degree C toluene 10ml were mixed for 0.25 hours, and 0.03g of tin octanoate was added. The generated copolymer was taken out after the 3-hour reaction.

[0113] The obtained lactic-acid system polyether polyester was transparent, and was number average molecular weight 87,000 and weight average molecular weight 150,000 from GPC. The glass transition point was observed by about 53 degrees C, and the melting point was about 158 degrees C. Moreover, \*\*\*\* fracture distortion was [ 490 kgf/cm<sup>2</sup> and the initial elastic modulus of \*\*\*\* of \*\*\*\* breaking strength ] 21,000 kgf/cm<sup>2</sup> 6%.

[0114] [Example 3] It taught 200ml separable flask, and under nitrogen-gas-atmosphere mind, it fused, polyether polyester (A-2) 30g obtained in the example 2, L-lactide 68g, D-lactide 2g, and 175-degree C toluene 10ml were mixed for 0.25 hours, and 0.03g of tin octanoate was added.

[0115] The generated copolymer was taken out after the 3-hour reaction. The obtained lactic-acid system polyether polyester was transparent, and was number average molecular weight 72,000 and weight average molecular weight 120,000 from GPC. Observed by about 53 degrees C although glass transition points were few, the melting point was about 158 degrees C. Moreover, \*\*\*\* fracture distortion was [ 420 kgf/cm<sup>2</sup> and the initial elastic modulus of \*\*\*\* of \*\*\*\* breaking strength ] 19,000 kgf/cm<sup>2</sup> 11%.

[0116] [Example 4] It taught 200ml separable flask, and under nitrogen-gas-atmosphere mind, it fused, polyether polyester (A-2) 50g obtained in the example 2, L-lactide 48g, D-lactide 2g, and 175-degree C toluene 10ml were mixed for 0.25 hours, and 0.03g of tin octanoate was added. The generated copolymer was taken out after the 3-hour reaction.

[0117] The obtained lactic-acid system polyether polyester was transparent, and was number average molecular weight 52,000 and weight average molecular weight 71,000 from GPC. Observed by about 53 degrees C although glass transition points were few, the melting point was about 153 degrees C. moreover, \*\*\*\* fracture distortion -- 200% and \*\*\*\* breaking strength -- 350 kgf/cm<sup>2</sup> and the initial elastic modulus of \*\*\*\* -- 6,200 kgf/cm<sup>2</sup> it was .

[0118] [Example 5] Heating stirring was carried out, teaching polypropylene-glycol 90g of number average molecular weight 3,000, ethylene glycol 2.0g, and 8.7g of sebacic acids to 300ml 4 opening flask which attached an agitator, a rectifier, and gas installation tubing, and carrying out a temperature up to it in 150 degrees C to 10 degrees C /under nitrogen-gas-atmosphere mind in an hour. Distilling off the water to generate, after checking that carried out the temperature up to 220 degrees C, and the distillate of water has stopped, titanium isopropyl titanate 0.01g was added and the deglycol reaction was performed at 220 degrees C under reduced pressure of 0.5Torr(s) for 3 hours.

[0119] it is [ polyester / which was obtained / polyether ] \*\*\*\*\* in number average molecular weight 19,000 and weight average molecular weight 32,000 -- it was liquefied. It taught 200ml separable flask, and under nitrogen-gas-atmosphere mind, it fused, obtained polyether polyester 30g, L-lactide 68g, D-lactide 2g, and 175-degree C toluene 10ml were mixed for 0.25 hours, and 0.03g of tin octanoate was added.

[0120] The generated copolymer was taken out after the 3-hour reaction. The obtained lactic-acid system polyether polyester was transparent, and was number average molecular weight 45,000 and

weight average molecular weight 88,000 from GPC. The glass transition point was observed by about 50 degrees C, and the melting point was about 156 degrees C. Moreover, \*\*\*\* fracture distortion was [ 360 kgf/cm<sup>2</sup> and the initial elastic modulus of \*\*\*\* of \*\*\*\* breaking strength ] 18,000 kgf/cm<sup>2</sup> 10%.

[0121] [Example 6] Heating stirring was carried out, teaching propylene glycol 26g and 50g of sebacic acids to 300ml 4 opening flask which attached an agitator, a rectifier, and gas installation tubing, and carrying out a temperature up to it in 150 degrees C to 10 degrees C /under nitrogen-gas-atmosphere mind in an hour. The temperature up was carried out to 220 degrees C, distilling off the water to generate, after checking that the distillate of water has stopped, titanium isopropyl titanate 0.01g was added and the deglycol reaction was performed at 220 degrees C under reduced pressure of 0.5Torr(s) for 3 hours.

[0122] it is [ polyester / which was obtained ] \*\*\*\*\* in number average molecular weight 17,000 and weight average molecular weight 28,000 -- it was liquefied. Obtained polyester 14g and polypropylene-glycol 60g of number average molecular weight 3,000 were taught to 200ml 4 opening flask which attached an agitator, a rectifier, and gas installation tubing, and the deglycol reaction was performed at 220 degrees C under reduced pressure of 0.5Torr for 3 hours. it is [ polyester / which was obtained / polyether ] \*\*\*\*\* in number average molecular weight 20,000 and weight average molecular weight 35,000 -- it was liquefied.

[0123] It taught 200ml separable flask, and under nitrogen-gas-atmosphere mind, it fused, obtained polyether polyester 30g, L-lactide 68g, D-lactide 2g, and 175-degree C toluene 10ml were mixed for 0.25 hours, and 0.03g of tin octanoate was added. The generated copolymer was taken out after the 3-hour reaction.

[0124] The obtained lactic-acid system polyether polyester was transparent, and was number average molecular weight 61,000 and weight average molecular weight 120,000 from GPC. The glass transition point was observed by about 53 degrees C, and the melting point turned out to be about 152 degrees C. moreover, \*\*\*\* fracture distortion -- 15% and \*\*\*\* breaking strength -- 330 kgf/cm<sup>2</sup> and the initial elastic modulus of \*\*\*\* -- 12,000 kgf/cm<sup>2</sup> it was .

[0125] [Example 7] Heating stirring was carried out, teaching polyethylene-glycol 110g of number average molecular weight 2,000, ethylene glycol 3.2g, and 16g of sebacic acids to 300ml 4 opening flask which attached an agitator, a rectifier, and gas installation tubing, and carrying out a temperature up to it in 150 degrees C to 10 degrees C /under nitrogen-gas-atmosphere mind in an hour.

[0126] The temperature up was carried out to 220 degrees C, distilling off the water to generate, after checking that the distillate of water has stopped, titanium isopropyl titanate 0.01g was added and the deglycol reaction was performed at 220 degrees C under reduced pressure of 0.5Torr(s) for 3 hours. The obtained polyether polyester was the solid-state of number average molecular weight 18,000 and weight average molecular weight 33,000.

[0127] It taught 200ml separable flask, and under nitrogen-gas-atmosphere mind, it fused, obtained polyether polyester 30g, L-lactide 68g, D-lactide 2g, and 175-degree C toluene 10ml were mixed for 0.25 hours, and 0.03g of tin octanoate was added. The generated copolymer was taken out after the 3-hour reaction.

[0128] The obtained lactic-acid system polyether polyester was transparent, and was number average molecular weight 45,000 and weight average molecular weight 88,000 from GPC. The glass transition point was observed by about 50 degrees C, and the melting point was about 156 degrees C. moreover, \*\*\*\* fracture distortion -- 120% and \*\*\*\* breaking strength -- 200 kgf/cm<sup>2</sup> and the initial elastic modulus of \*\*\*\* -- 7,100 kgf/cm<sup>2</sup> it was .

[0129] [Example 8] Heating stirring was carried out, teaching polyethylene-glycol 140g of number average molecular weight 20,000, ethylene glycol 1.0g, and 2.0g of sebacic acids to 300ml 4 opening flask which attached an agitator, a rectifier, and gas installation tubing, and carrying out a temperature up to it in 150 degrees C to 10 degrees C /under nitrogen-gas-atmosphere mind in an hour.

[0130] The temperature up was carried out to 220 degrees C, distilling off the water to generate, after checking that the distillate of water has stopped, titanium isopropyl titanate 0.01g was added and the deglycol reaction was performed at 220 degrees C under reduced pressure of 0.5Torr(s) for 3 hours. The

obtained polyether polyester was the solid-state of number average molecular weight 42,000 and weight average molecular weight 58,000.

[0131] It taught 200ml separable flask, and under nitrogen-gas-atmosphere mind, it fused, obtained polyether polyester 30g, L-lactide 68g, D-lactide 2g, and 175-degree C toluene 10ml were mixed for 0.25 hours, and 0.03g of tin octanoate was added. The generated copolymer was taken out after the 3-hour reaction.

[0132] The obtained lactic-acid system polyether polyester was transparent, and was number average molecular weight 72,000 and weight average molecular weight 130,000 from GPC. The glass transition point was observed by about 50 degrees C, and the melting point was about 152 degrees C. moreover, \*\*\*\* fracture distortion -- 94% and \*\*\*\* breaking strength -- 210 kgf/cm<sup>2</sup> and the initial elastic modulus of \*\*\*\* -- 6,800 kgf/cm<sup>2</sup> it was .

[0133] [Example 9] Heating stirring was carried out, teaching ethylene glycol 57g and 100g of adipic acids to 300ml 4 opening flask which attached an agitator, a rectifier, and gas installation tubing, and carrying out a temperature up to it in 150 degrees C to 10 degrees C /under nitrogen-gas-atmosphere mind in an hour. The temperature up was carried out to 200 degrees C, distilling off the water to generate, and the deglycol reaction was performed for 30 minutes at 200 degrees C under reduced pressure of 3Torr after checking that the distillate of water has stopped.

[0134] Polypropylene-glycol 714g of number average molecular weight 3,000 and titanium isopropyl titanate 0.01g were added to this, and the deglycol reaction was performed to it at 200 degrees C under reduced pressure of 0.5Torr(s) for 3 hours. it is [ polyester / (A-3) / which was obtained / polyether ] \*\*\*\*\* in number average molecular weight 25,000 and weight average molecular weight 40,000 - it was liquefied.

[0135] It taught 200ml separable flask, and under nitrogen-gas-atmosphere mind, it fused, obtained polyether polyester 30g, L-lactide 68g, D-lactide 2g, and 175-degree C toluene 10ml were mixed for 0.25 hours, and 0.03g of tin octanoate was added. The generated copolymer was taken out after the 3-hour reaction.

[0136] The obtained lactic-acid system polyether polyester was transparent, and was number average molecular weight 610,000 and weight average molecular weight 11,000 from GPC. The glass transition point was observed by about 50 degrees C, and the melting point was about 152 degrees C. Moreover, \*\*\*\* fracture distortion was [ 280 kgf/cm<sup>2</sup> and the initial elastic modulus of \*\*\*\* of \*\*\*\* breaking strength ] 10,000 kgf/cm<sup>2</sup> 27%.

[0137] [Example 10] Heating stirring was carried out, teaching ethylene glycol 71g and 100g of amber acids to 3l. 4 opening flask which attached an agitator, a rectifier, and gas installation tubing, and carrying out a temperature up to it in 150 degrees C to 10 degrees C /under nitrogen-gas-atmosphere mind in an hour. The temperature up was carried out to 200 degrees C, distilling off the water to generate, and the deglycol reaction was performed at 200 degrees C under reduced pressure of 3Torr after checking that the distillate of water has stopped for 30 minutes.

[0138] Polypropylene-glycol 890g of number average molecular weight 3,000 and titanium isopropyl titanate 0.01g were added to this, and the deglycol reaction was performed to it at 200 degrees C under reduced pressure of 0.5Torr(s) for 3 hours. it is [ polyester / which was obtained ] \*\*\*\*\* in number average molecular weight 22,000 and weight average molecular weight 39,000 -- it was liquefied.

[0139] It taught 200ml separable flask, and under nitrogen-gas-atmosphere mind, it fused, obtained polyether polyester 30g, L-lactide 68g, D-lactide 2g, and 175-degree C toluene 10ml were mixed for 0.25 hours, and 0.03g of tin octanoate was added. The generated copolymer was taken out after the 3-hour reaction.

[0140] The obtained lactic-acid system polyether polyester was transparent, and was number average molecular weight 720,000 and weight average molecular weight 12,000 from GPC. The glass transition point was observed by about 50 degrees C, and the melting point turned out to be about 152 degrees C. Moreover, \*\*\*\* fracture distortion was [ 270 kgf/cm<sup>2</sup> and the initial elastic modulus of \*\*\*\* of \*\*\*\* breaking strength ] 10,000 kgf/cm<sup>2</sup> 29%.

[0141] [Example 11] Polyether polyester (A-3) 30g obtained in the example 9 and 0.18g (it abbreviates to PMDA henceforth) of pyromellitic dianhydride were taught to 300ml 4 opening flask which attached an agitator, a rectifier, and gas installation tubing, and the deglycol reaction was performed at 200 degrees C under reduced pressure of 0.5Torr for 3 hours. it is [ polyester / which was obtained ] \*\*\*\*\* in number average molecular weight 33,000 and weight average molecular weight 90,000 - it was liquefied.

[0142] It taught 200ml separable flask, and under nitrogen-gas-atmosphere mind, it fused, obtained polyether polyester 30g, L-lactide 68g, D-lactide 2g, and 175-degree C toluene 10ml were mixed for 0.25 hours, and 0.03g of tin octanoate was added. The generated copolymer was taken out after the 3-hour reaction.

[0143] The obtained lactic-acid system polyether polyester was transparent, number average molecular weight 65,000, weight average molecular weight 13,000, and a glass transition point were observed by about 53 degrees C from GPC, and the melting point was about 150 degrees C. Moreover, \*\*\*\* fracture distortion was [ 220 kgf/cm<sup>2</sup> and the initial elastic modulus of \*\*\*\* of \*\*\*\* breaking strength ] 9,100 kgf/cm<sup>2</sup> 43%.

[0144] [Example 12] Heating stirring was carried out, teaching ethylene glycol 57g, 100g of adipic acids, and 0.6g of pyromellitic dianhydride to 300ml 4 opening flask which attached an agitator, a rectifier, and gas installation tubing, and carrying out a temperature up to it in 150 degrees C to 10 degrees C /under nitrogen-gas-atmosphere mind in an hour. The temperature up was carried out to 200 degrees C, distilling off the water to generate, and the deglycol reaction was performed at 200 degrees C under reduced pressure of 3Torr after checking that the distillate of water has stopped for 30 minutes.

[0145] Polypropylene-glycol 719g of number average molecular weight 3,000 and titanium isopropyl titanate 0.01g were added to this, and the deglycol reaction was performed to it at 200 degrees C under reduced pressure of 0.5Torr(s) for 3 hours. it is [ polyester / which was obtained / polyether ] \*\*\*\*\* in number average molecular weight 25,000 and weight average molecular weight 40,000 - it was liquefied.

[0146] It taught 200ml separable flask, and under nitrogen-gas-atmosphere mind, it fused, obtained polyether polyester 30g, L-lactide 68g, D-lactide 2g, and 175-degree C toluene 10ml were mixed for 0.25 hours, and 0.03g of tin octanoate was added. The generated copolymer was taken out after the 3-hour reaction. The obtained lactic-acid system polyether polyester was transparent, and was number average molecular weight 51,000 and weight average molecular weight 110,000 from GPC. The glass transition point was observed by about 50 degrees C, and the melting point was about 152 degrees C. Moreover, \*\*\*\* fracture distortion was [ 330 kgf/cm<sup>2</sup> and the initial elastic modulus of \*\*\*\* of \*\*\*\* breaking strength ] 10,000 kgf/cm<sup>2</sup> 31%.

[0147] [Example 13] 80g (PE[ by Sanyo Chemical Industries, Ltd. ]- 75) of block copolymers of the polyethylene glycol and polypropylene glycol of polyester (A-1) 13g and number average molecular weight 4,025 obtained in the example 2 was taught to 200ml 4 opening flask which attached an agitator, a rectifier, and gas installation tubing, and the deglycol reaction was performed at 220 degrees C under reduced pressure of 0.5Torr for 3 hours. it is [ polyester / which was obtained / polyether ] \*\*\*\*\* in number average molecular weight 25,000 and weight average molecular weight 44,000 -- it was liquefied.

[0148] It taught 200ml separable flask, and under nitrogen-gas-atmosphere mind, it fused, obtained polyether polyester 30g, L-lactide 68g, D-lactide 2g, and 175-degree C toluene 10ml were mixed for 0.25 hours, and 0.03g of tin octanoate was added. The generated copolymer was taken out after the 3-hour reaction.

[0149] The obtained lactic-acid system polyether polyester was transparent, number average molecular weight 57,000, weight average molecular weight 83,000, and a glass transition point were observed by about 50 degrees C from GPC, and the melting point was about 152 degrees C. moreover, \*\*\*\* fracture distortion -- 32% and \*\*\*\* breaking strength -- 310 kgf/cm<sup>2</sup> and the initial elastic modulus of \*\*\*\* -- 10,000 kgf/cm<sup>2</sup> it was .

[0150] [Example 14] Polyester (A-1) 11g obtained in the example 2, 90g (PE[ by Sanyo Chemical

Industries, Ltd. ]- 75) of block copolymers of the polyethylene glycol and polypropylene glycol of number average molecular weight 4,025, 10g (GL[ by Sanyo Chemical Industries, Ltd. ]- 3000) of triol mold block copolymers of the polyethylene glycol and polypropylene glycol of number average molecular weight 3,000 An agitator, It taught 200ml 4 opening flask which attached a rectifier and gas installation tubing, and the deglycol reaction was performed at 220 degrees C under reduced pressure of 0.5Torr for 3 hours.

[0151] it is [ polyester / which was obtained / polyether ] \*\*\*\*\* in number average molecular weight 31,000 and weight average molecular weight 59,000 -- it was liquefied. It taught 200ml separable flask, and under nitrogen-gas-atmosphere mind, it fused, obtained polyether polyester 30g, L-lactide 68g, D-lactide 2g, and 175-degree C toluene 10ml were mixed for 0.25 hours, and 0.03g of tin octanoate was added. The generated copolymer was taken out after the 3-hour reaction.

[0152] The obtained lactic-acid system polyether polyester was transparent, and was number average molecular weight 79,000 and weight average molecular weight 130,000 from GPC. The glass transition point was observed by about 51 degrees C, and the melting point was about 152 degrees C. Moreover, \*\*\*\* fracture distortion was [ 290 kgf/cm<sup>2</sup> and the initial elastic modulus of \*\*\*\* of \*\*\*\* breaking strength ] 10,000 kgf/cm<sup>2</sup> 40%.

[0153] The hotpress was used for the lactic-acid system copolymerization polyether polyester obtained in the example 3, and the sheet (thickness 10cmx10cm and 100 micrometers) was created. The weight average molecular weight of the sheet obtained under 165 degrees C, 200kg/cm<sup>2</sup>, and the press conditions for 2 minutes was 117,000. This sheet was laid underground into soil and the biodegradation test was tried. A result is shown in Table 1.

[0154]

[Table 1]

浸漬日数	重量平均分子量	外観
0日	117,000	剛直 透明
30日	52,000	剛直 半透明
100日	15,000	脆い 白色
200日	6,200	脆い 白色

[0155] [Example 1 of a comparison] Toluene 15ml was added to L-lactide 100g, under nitrogen-gas-atmosphere mind, the dissolution and mixing of were done, 0.03g of tin octanoate was added as a ring-opening-polymerization catalyst, and 175 degrees C of polymerizations were performed for 3 hours for 0.25 hours. Toluene was removed under reduced pressure after the reaction. Generated L-poly(lactic acid) was transparent and colorless resin of weight average molecular weight 273,000 and number average molecular weight 140,000. The glass transition point of this was about 57 degrees C, and the melting point was about 158 degrees C. moreover, \*\*\*\* fracture distortion -- 3% and \*\*\*\* breaking strength -- 500 kgf/cm<sup>2</sup> and the initial elastic modulus of \*\*\*\* -- 16,000 kgf/cm<sup>2</sup> it was .

[0156] [the example 2 of a comparison] -- L-lactide 70g -- poly caprolactone (product made from UCC "tone") 30g, and toluene 15g -- in addition, under nitrogen-gas-atmosphere mind, the dissolution and mixing of were done, 175 degrees C of 0.03g of tin octanoate were added as a ring-opening-polymerization catalyst for 0.25 hours, and the polymerization was performed for 3 hours. Toluene was removed under reduced pressure after the reaction. The generated lactide system copolymerization polyether polyester was resin of the white of weight average molecular weight 223,000 and number average molecular weight 110,000. For about 47 degrees C and the melting point, about 149 degrees C and \*\*\*\* fracture distortion were [ the glass transition point of this / 250 kgf/cm<sup>2</sup> and the initial elastic modulus of \*\*\*\* of \*\*\*\* breaking strength ] 13,000 kgf/cm<sup>2</sup> 30%.

[0157] The measurement result about the lactic-acid system copolymer obtained in the example and the



example of a comparison is indicated from Table 2 to a front table. In addition, the abbreviation of front Naka shows the following contents.

[0158] sebacic-acid: -- SeA adipic-acid: -- AA succinic-acid: -- SA pyromellitic dianhydride :P MDA

[0159] Ethylene glycol: EG propylene glycol :P G polyethylene glycol :P EG polypropylene glycol :P

The triol mold block copolymer of PGPEG, block-copolymer:EbPPEG which is PPG, and PPG: tEbP

[0160]

[Table 2]

項 目		実施例 1	実施例 2	実施例 3	実施例 4
L-LD (重量%)		68	85	68	48
D-LD (重量%)		2	5	2	2
ポリ エ ー ス テ ル	組成	SeA/EG/PPG	SeA/EG/PPG	SeA/EG/PPG	SeA/EG/PPG
	組成比(モル%)	100/10/90	100/75/35	100/75/35	100/75/35
	ポリエーテルポリオール のMw	400	3000	3000	3000
	量(重量%)	30	10	30	50
	Mw $\times 10^{-3}$	33	35	35	35
Mw $\times 10^{-3}$		93	150	120	71
ガラス転移点(℃)		50	53	53	53
融点(℃)		156	157	158	153
引張破断歪(%)		10	6	11	200
引張破断強度 (kgf/cm <sup>2</sup> )		390	490	420	350
引張初期弾性率 (kgf/cm <sup>2</sup> )		13000	22000	19000	6200

[0161]

[Table 3]

項 目		実施例 5	実施例 6	実施例 7	実施例 8
L-LD (重量%)		68	68	68	68
D-LD (重量%)		2	2	2	2
ポリ エー テ ル エ ス テ ル	組成	SeA/EG/PPG	SeA/PG/PPG	SeA/EG/PEG	SeA/EG/PEG
	組成比(モル%)	100/30/70	100/75/35	100/30/70	100/30/70
	ポリエーテルジオールのMw	3000	3000	2000	20000
	量(重量%)	30	30	30	30
	Mw $\times 10^{-3}$	32	35	33	58
Mw $\times 10^{-3}$		88	120	88	130
ガラス転移点(℃)		50	53	50	50
融点(℃)		156	152	156	152
引張破断歪(%)		10	15	120	94
引張破断強度 (kgf/cm <sup>2</sup> )		360	330	200	210
引張初期弾性率 (kgf/cm <sup>2</sup> )		18000	12000	7100	6800

[0162]

[Table 4]

項 目		実施例 9	実施例 1 0	実施例 1 1	実施例 1 2
L-LD (重量%)		6 8	6 8	6 8	6 8
D-LD (重量%)		2	2	2	2
ポリ エー テ ル ポ リ エ ス テ ル	組成	AA/EG/PPG	SA/EG/PPG	AA/EG/PPG /PMDA	AA/EG/PPG /PMDA
	組成比(モル%)	100/75/35	100/75/35	100/75/35	100/75/35
	ポリエーテルポリアールのMw	3 0 0 0	3 0 0 0	3 0 0 0	3 0 0 0
	量 (重量%)	3 0	3 0	3 0	3 0
	Mw $\times 10^{-3}$	4 0	3 9	9 0	4 0
Mw $\times 10^{-3}$		1 1 0	1 2 0	1 3 0	1 1 0
ガラス転移点 (°C)		5 0	5 0	5 3	5 0
融点 (°C)		1 5 2	1 5 2	1 5 0	1 5 2
引張破断歪 (%)		2 7	2 9	4 3	3 1
引張破断強度 (kgf/cm <sup>2</sup> )		2 8 0	2 7 0	2 2 0	3 3 0
引張初期弾性率 (kgf/cm <sup>2</sup> )		1 0 0 0 0	1 0 0 0 0	9 1 0 0	1 0 0 0 0

[0163]

[Table 5]

項 目		実施例 1 3	実施例 1 4
L-LD (重量%)		6 8	6 8
D-LD (重量%)		2	2
ポリ エー ステ ル	組成	SeA/EG/EbP	SeA/EG/EbP/tEbP
	組成比(モル%)	100/75/35	100/47/ 46/7
	ポリエーテルのMw	4 0 2 5	EbP : 4 0 2 5 tEbP : 3 0 0 0
	量 (重量%)	3 0	3 0
	Mw $\times 10^{-3}$	4 4	5 9
Mw $\times 10^{-3}$		8 3	1 3 0
ガラス転移点 (°C)		5 0	5 1
融点 (°C)		1 5 2	1 5 2
引張破断歪 (%)		3 2	4 0
引張破断強度 (kgf/cm <sup>2</sup> )		3 1 0	2 9 0
引張初期弾性率 (kgf/cm <sup>2</sup> )		1 0 0 0 0	1 0 0 0 0

[0164]

[Table 6]

項 目		比較例 1	比較例 2
原	L-LD (wt%)	1 0 0	7 0
	D-LD (wt%)	0	0
料	ポリエステル	—	トーン
	(wt%)	—	3 0
	Mw×10 <sup>-3</sup>	—	—
Mw×10 <sup>-3</sup>		2 7 3	2 2 3
ガラス転移点 (℃)		5 7	4 7
融点 (℃)		1 5 8	1 4 9
引張破断歪 (%)		3	3 0
引張破断強度 (kgf/cm <sup>2</sup> )		5 0 0	2 5 0
引張初期弾性率 (kgf/cm <sup>2</sup> )		1 6 0 0 0	1 3 0 0 0

[0165]

[Translation done.]